ON SEQUENTIAL DESIGN OF EXPERIMENTS IN MULTIRESPONSE MODEL DISCRIMINATION

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CERTIFICATE

It is certified that the work entitled 'On sequential design of experiments in multiresponse Model discrimination ' has been carried out under my supervision and has not submitted elsewhere for a degree.

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(S. GANGULY)
Author

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NOMENCLATURE

A	Chemical species A in the reaction mixture.	
B	Chemical species B in the reaction mixture.	
c	Variable constant defined by Eq (2.2.18)	
C	Chemical species C in the reaction mixture.	
$\underset{\sim}{C}$	Symmetric positive definite matrix defined by Eq.(2.1.4)	(uxu)
D	Objective function to be maximized in Roth's criterion	
$\underline{e}$	Vector of random normal deviates following N(0,1) distribution	(uX1)
E(.)	Expected value of (.)	
$f_{rj}(\underline{x}^q_{rj},\theta_r)$	Expected value of the j^{th} response for the r^{th} model and q^{th} experimental setting.	
f_r	Vector of the expected values of observations for the r^{th} model.	(uX1)
f	Vector of the expected values of observations for the models.	(vuX1)
f^i_{ql}	i^{th} response for the l^{th} model and q^{th} experimental setting.	
f_{qk}	1st derivative of j^{th} response function w.r.t. θ_k evaluated at $\underline{x}^q_{rj}$.	
$\underline{F}_j(\underline{x},\underline{\theta})$	$(f_{rj}(\underline{x}^1_{rj}, \underline{\theta}_r) \; \ldots\ldots \; f_{rj}(\underline{x}^N_{rj}, \underline{\theta}_r))^T$	(NX1)
$\underline{F}$	$(\underline{F}(\underline{x}, \underline{\theta}) \; \ldots\ldots \; F_u(\underline{x}, \underline{\theta}))^T$	(NuX1)
F_j'	matrix whose elements are f_{qk}	(NXm)
$\underline{\underline{F}}'$	$(\underline{\underline{F}}'_1 \; \underline{\underline{F}}'_2 \; \ldots\ldots \; \underline{\underline{F}}'_v)^T$	

g_k	lower constraint on the independent variables	(N9Xm)
h_k	upper constraint on the independent variable	
i	Subscript/superscript referring to i^{th} response Subscript referring to the i^{th} independent variable in Box's optimization algorithm defined Eq (2.4.3)	
I(r:s)	Expected information in favour of choosing model r over model s	
j	Subscript/superscript referring to j^{th} response subscript referring to the vertex in Box's optimization algorithm defined in equation (2.4.3)	
J(r:s)	Kullbacks criterion for distinguishing between two states of nature r and s.	
k	subscript denoting parameter referring to 'complex points' in Box's optimization algorithm	
K_v	Scalar discriminant function in Box and Hill's criterion	
l	referring to the l^{th} model	
$L(\underline{\theta})$	Likelihood of the parameters	
L_r	Likelihood of the r^{th} model	
m	number of parameters	
n	number of indepent variables	
N	number of experiments	
N*	new experiments to be conducted	
P_r^N	Probability of the r^{th} model after N runs	
q	referring to experiments	
r	referring to models	
r_{ij}	random numbers	

s	referring to models.
	Experiment in Eq(2.3.2)
t	time
u	Total number of responses
v	total number of models
$\underline{x}$	vector of independent variables
x_1	concentration of component A
x_2	concentration of component B
x_3	concentration of component C
$\underline{\underline{X}}_{rj}$	matrix of partial derivatives defined by Eq(2.3.7) (Nxm)
$\underline{\underline{X}}_r^{N+1}$	matrix of partial derivatives defined by Eq(2.3.10) (uxm)
$\underline{\underline{X}}$	Design matrix for linear multiresponse model (Nxm)
$\underline{Y}, y$	vector of responses

Greek symbols

α	Reflection factor
β	convergence criterion
γ	number of iteration
δ	Reset value
$\underline{\delta}$	Increment in $\underline{\theta}_o$ as given by Eq (2.2.16)
ε_{jq}	Experimental error associated with j^{th} response (ux1)
σ_{ij}	is the (i,j) element of the variance covariance matrix
$\underline{\theta}$	vector of parameters (mx1)
$\underline{\underline{\Sigma}}$	variance covariance matrix (uxu)

Superscript/subscript

i,j	for responses
q	for experiment
r,s	for models
$\hat{}$	for estimates

CHAPTER 1

INTRODUCTION:

In any scientific study, proper description of the physical or chemical phenomenon forms an important part of the work. These phenomena can generally be expressed through mathematical models showing the relationship between the dependent and the independent variables. One expects that these models should be unique and which implies that only one model should be sufficient to describe a particular phenomenon. Unfortunately the experimenter observes, specially in complicated cases, that a phenomenon can be described by more than one model. This is mainly because the mechanisms, on which the models are based, are seldom perfectly known and consequently the experimenter faces the problem of identifying the most adequate model.

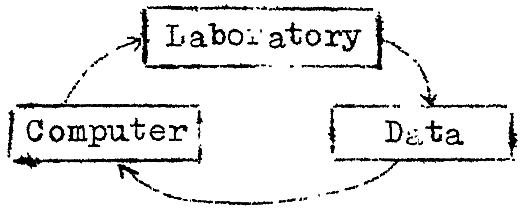
For example in the case of non-newtonian fluid flow one can obtain shear stress vs. shear rate data which when fitted to the different available fluid models e.g. Bingham plastic, Rheopectic, Thixotropic etc., could give apparently meaningful results in each case. Under this circumstances one has to properly classify the fluid, rather one has to identify the most adequate model.

The number of dependent variables in the fluid flow problem, explained above, is only one and is called a single response system. In chemical kinetics where more than one component is present during the reaction and observations are made on all the components the number of dependent variables is more than one. In such a case the system is called a multiresponse one.

While studying heterogeneous chemical reaction which is a multiresponse situation, where most of the mechanisms have not yet been properly identified, one can propose more than one mechanistic model to describe the reacting system. For example while using Hougen and Watson's method of approach the main steps involved are (i) Adsorption of the components on the surface (ii) Reaction of the adsorbed components on the surface (iii) Desorption of the components from the surface.

Considering any of the above step as the rate controlli step one can propose more than one mechanistic model for the same chemical reaction. These examples and many other which have not been explained necessitates an ellaborate study of th methods for model discrimination. Also the use of multirespon techniques for parameter estimation and model discrimination not only increases the precision of the estimates of parameter but also reduces the volume of confidence ellipsoid.

The strategy of model discrimination and parameter estimation can be described schematically as given below,



An experimenter does his experiment in the laboratory which gives information in the form of data. These data are used for parameter estimation and discrimination among rival models through computer. The iterative process can be carried on until a satisfactory level is reached.

LITERATURE SURVEY:

Extensive literature is available on parameter estimatic and model discrimination for single response models but for multiresponse situations the technique available for both parameter estimation and model discrimination seem to be rather limited which are discussed briefly in the following sections.

Ray[1], Singh and Rao[2] have reviewed the techniques available for parameter estimation in multiresponse situations. Box and Draper[3] discussed the Bayesian estimation of parameters. Draper and Hunter[4,5] dealt with design of experimen for parameter estimation in multiresponse cases. Beauchamp and Cornell[6] mathematically converted multiresponse model

to single response one and gave a novel technique for simultaneous non linear estimation of parameters. Hunter[7] reviewed different criteria for estimation of unknown constant from multiresponse data. Mezaki and Butt[8] proved that Bayesian estimation of parameters is far better than generalized non linear least square technique. M.J. Box[9,10,11] used the concept of non constant variance covariance matrix for improved estimate of parameters. Box, et.al.[12] discussed some problems associated with analysis of multiresponse data.

In model discrimination the same criterion used for discrimination in single response cases can be used for multiresponse cases with proper modifications. These method comprise mainly of

i) Likelihood methods
ii) Bayesian methods

Likelihood methods

In these methods certain error structure is assumed a priori and based on this the likelihood of a particular model is calculated. The likelihood ratios, defined as the ratios of maximum likelihood of a given model to that of various other models are calculated and based on a likelihood ratio of 100 or more, discrimination is achieved. This method has been amply exemplified by Rao, et.al.[13] in an earlier study.

Bayesian Methods

In the case of Bayesian methods, certain prior probabilities are assumed for the models and in the light of the data the model probabilities are updated resulting in the posterior probabilities. Invariably Bayes theorem is used for calculating the posterior probabilities for the different models as given by the relation

$$P_r^{N+1} = \frac{P_r^N \; L_r}{\sum_{r=1}^{V} P_r^N \; L_r} \tag{1.1}$$

where P_r^{N+1} is the posterior probability for the rth model after (N+1) runs and P_r^N is the prior probability for the rth model before the (N+1) run. L_r is the likelihood for the rth model and V represents the total number of models considered.

Also a knowledge of error structure is required for the calculation of likelihood and certain prior probabilities are to be assumed for each of the competing models. In the absence of any prior knowledge equal prior probabilities are assumed for all the competing models. Model discrimination is achieved when the posterior probability of any one of the models exceeds a preset value.

Ray[1] suggested a novel technique for model discrimination by extending the parameter estimation criterion of Box and Draper[3]. The method though appealing failed to discriminate properly in some cases.

Sequential design for discrimination

After conducting N experimental trials the posterior probabilities of the different competing models are calculated as described in earlier section. If the discrimination is not achieved additional experimental runs are necessary and the experiments are conducted at such conditions of the independent variables which will give the best discrimination amongst the rival models. The criterion available for the sequential design of experiments are briefly discussed in the following sections:

Roth's[14] criterion:

For sequential design of experiments Roth suggested that the experiments be conducted at appropriate condition of independent variables which maximizes the objective function D given by

$$D = \prod_{i=1}^{U} \left(\sum_{r=1}^{V} \left(P_r \prod_{l=1}^{V} \left| \hat{f}_{ql}^{\,i} - \hat{f}_{qr}^{\,i} \right| \right) \right) \qquad (1.2)$$

where i represents the responses, r represents the models and q represents the qth setting of experimental conditions. P_r represents the prior probability of the rth model before the (N+1) experiment and $\hat{f}_{ql}$ represents the predicted value of the ith response for the lth model and kth experimental setting.

Box and Hill's[15,16] criterion

The most comprehensive technique that has so far been described is by Box and Hill.

They suggested that the following objective function be maximized

$$K_V = \frac{1}{2} \sum_{r=1}^{V} \sum_{s=r+1}^{V} P_r^N P_s^N \Big[\text{trace}\Big\{ \Sigma_r^{N+1} (\Sigma_s^{N+1})^{-1} + \Sigma_s^{N+1} \cdot (\Sigma_r^{N+1})^{-1} - 2I_u \Big\} + (\hat{Y}_r^{N+1} - \hat{Y}_s^{N+1})^T \Big\{ (\Sigma_s^{N+1})^{-1} + (\Sigma_r^{N+1})^{-1} \Big\} (\hat{Y}_r^{N+1} - \hat{Y}_s^{N+1}) \Big] \qquad (1.3)$$

where all the terms and the underlying theory have been discussed in details in Chapter 2.

Hsiang and Reilly[17] by suitable assumptions subverted Box and Hill's assumptions of normality, known variance and linearity of models in the neighbourhood of parameter estimates. Prasad and Rao[18] for the first time used the expected likelihood instead of point likelihood in Box and Hill's criterion. They applied the modification to study catalyst fouling system and proved that the rate of discrimination was much faster compared to the original Box and Hill's approach.

OBJECTIVE OF THE PRESENT STUDY:

As seen from the above literature survey the parameter estimation and model discrimination in multiresponse situations pose no special difficulties. However the effective use of sequential design of experiments has not been practically demonstrated barring for the only study by Prasad and Rao[18]. In the present investigation it is proposed to see how effective is the sequential design of experiments for discrimination, between rival models in multiresponse situations, proposed by Box and Hill[15,16]. Various illustrative examples are considered to see how the posterior probabilities vary during the course of the sequential design of experimentation.

CHAPTER 2
THEORY

2.1 Data Generation:

In order to test the model discrimination criterion[15,16] for multiresponse case with unknown variance covariance matrix, with reference to Chemical Engineering problems, a chemical reaction was considered for the present study.

A reacting system comprising of components A,B, and C undergoing a chemical reaction represents a general chemical reaction and some assumptions were imposed on the state of the reaction so that the study could be focussed on model discrimination rather than on the details of the kinetic reactions.

The assumptions are

(1) The reaction is carried out in a batch reactor.
(2) The reaction is isothermal.
(3) There is no change in total moles, volume and density.
(4) The reaction follows first order kinetics.

The possible competitive models suggested are

$$\begin{aligned}
&\text{(i)} \quad A \xrightarrow{\theta_{11}} B \underset{\theta_{13}}{\overset{\theta_{12}}{\rightleftharpoons}} C \\
&\text{(ii)} \quad A \underset{\theta_{22}}{\overset{\theta_{21}}{\rightleftharpoons}} B \xrightarrow{\theta_{23}} C \\
&\text{(iii)} \quad A \xrightarrow{\theta_{31}} B \xrightarrow{\theta_{32}} C \\
&\text{(iv)} \quad A \underset{\theta_{42}}{\overset{\theta_{41}}{\rightleftharpoons}} B \underset{\theta_{44}}{\overset{\theta_{43}}{\rightleftharpoons}} C
\end{aligned} \qquad (2.1)$$

where θ are the rate constants or parameters and all the four assumptions made, holds good for these models.

The rate expressions for the models considered are

<u>Model 1</u> $\quad A \xrightarrow{\theta_{11}} B \underset{\theta_{13}}{\overset{\theta_{12}}{\rightleftharpoons}} C$

$$Y_{11} = \frac{d\ x_1}{dt} = \theta_{11}\ x_1$$

$$Y_{12} = \frac{d\ x_2}{dt} = \theta_{11}\ x_1 - \theta_{12}\ x_2 + \theta_{13}\ x_3$$

$$Y_{13} = \frac{d\ x_3}{dt} = \theta_{12}\ x_2 - \theta_{13}\ x_3$$

<u>Model 2</u> $\quad A \underset{\theta_{22}}{\overset{\theta_{21}}{\rightleftharpoons}} B \xrightarrow{\theta_{23}} C$

$$Y_{21} = \frac{d\ x_1}{dt} = -\ \theta_{21}\ x_1$$

$$Y_{22} = \frac{d\ x_2}{dt} = \theta_{21}\ x_1 - \theta_{22}\ x_2 - \theta_{23}\ x_2$$

$$Y_{23} = \frac{d\ x_3}{dt} = \theta_{23}\ x_2$$

<u>Model 3</u> $\quad A \xrightarrow{\theta_{31}} B \xrightarrow{\theta_{32}} C$

$$Y_{31} = \frac{d\ x_1}{dt} = -\ \theta_{31}\ x_1$$

$$Y_{32} = \frac{d\ x_2}{dt} = \theta_{31}\ x_1 - \theta_{32}\ x_2$$

$$Y_{33} = \frac{d\ x_3}{dt} = \theta_{32}\ x_2$$

$$A \underset{\theta_{42}}{\overset{\theta_{41}}{\rightleftharpoons}} B \underset{\theta_{44}}{\overset{\theta_{43}}{\rightleftharpoons}} C$$

<u>Model 4</u>

$$Y_{41} = \frac{d\ x_1}{dt} = -\ \theta_{41}\ x_1 + \theta_{42}\ x_2$$

$$Y_{42} = \frac{d\ x_2}{dt} = \theta_{41}\ x_1 - \theta_{42}\ x_2 - \theta_{43}\ x_2 + \theta_{44}\ x_3$$

$$Y_{43} = \frac{d\ x_3}{dt} = \theta_{43}\ x_2 - \theta_{44}\ x_3$$

where Y represents response, x represents concentration of components A,B,C and θ represents rate constants or parameters.

Due to the non availability of experimental data, at the time of this study, efforts were made to get simulated data conforming with the assumptions already made. The following represents the scheme adopted to help overcoming the above mentioned deficiency:

(1) (A) Data were generated using model $A \rightarrow B \rightarrow C$

(i) without correlation among responses,
(ii) with correlation among responses.

(B) Data were generated using model $A \rightarrow B \rightleftharpoons C$

(i)without correlation among responses,
(ii)with correlation among responses.

(2) Data reported by (19) were considered.

A comprehensive survey about generation of data has been reported by Ray[1] but the data reported by Ray corresponds only to case 1B(i) and was also unrealistic in nature. In the

present study extensive data were generated corresponding to (1), compatible with the underlying theory given by earlier workers[1] representing real case study.

Considering the general multiresponse model

$$Y_{rj}^{q} = f_{rj} \left(\underline{x}_{rj}^{q} , \underline{\theta}_{r} \right) + \epsilon_{jq}$$

the data were generated after assigning some realistic values to θ.[20] The method consists of calculating the $\underline{x}$ values from the analytical expression for $\underline{x}$ vs t for the particular model considered[1] and at regular intervals of time 't' for the assumed set of parameter. Thereafter f_{rj} was calculated from the rate expression and finally the normal error was added to give the simulated response vector. To avoid the dependency in the expected values of responses[12] only (Y_{r2}, Y_{r3}) were taken as the response vector as we have from material balance

$$E(Y_{r1}) + E(Y_{r2}) + E(Y_{r3}) = 0 \qquad (2.1.1)$$

Also ϵ_{jq}, vector of errors for the qth experiment was assumed to have normal distribution, i.e.,

$$E(\underline{\epsilon}_q)=\underline{0} \text{ and } E(\underset{(u\times1)(1\times u)}{\underline{\epsilon}_q, \underline{\epsilon}_q^T}) = \underset{(u\times u)}{\underline{\underline{\Sigma}}^q} \qquad (2.1.2)$$

Since the elements of $\underline{\epsilon}_q$ are jointly normal for simulation the expression can be expressed[1] as a linear combination of independent random variables

$$\underset{(u\times 1)}{\underline{\epsilon}_q} = \underset{(u\times u)}{\underline{\underline{C}}^q} \cdot \underset{(u\times 1)}{\underline{e}} \tag{2.1.3}$$

where $\mathbf{C}^q$ is a non singular matrix

such that $\underline{\underline{\Sigma}}^q = \underline{\underline{C}}^q \underline{\underline{C}}'^q$ (2.1.4)

and elements of $\underline{e}$ has a N (0,1) distribution obtained from table of random normal deviates. While considering reaction kinetics study one inevitably faces the problem of choosing the response vector and the independent or controlled variable vector. An experimenter actually observes the concentrations of the components at regular intervals of time and it seems that it is more realistic to consider $(x_1, x_2, x_3)^T$ as the response vector and $(x_1^o, x_2^o, x_3^o, t)^T$ as the controlled vector where the superscript o indicates initial state and 't' represents time. Under this circumstance it may be justified to assume that the error of observation will follow a multivariate normal distribution. But it has been pointed out by Ray[1] that this consideration will not only make the system more complex but also may lead to noncovergence as far as parameter estimation is concerned.

The other alternative is to consider the rate vector $(\frac{d\,x_1}{dt}, \frac{d\,x_2}{dt}, \frac{d\,x_3}{dt})'$ as the response vector and concentration

vector $(x_1, x_2, x_3)^T$ as the controlled variable vector. But since rates are generally not experimentally measurable quantities rather these are calculated from concentration vs. time data, the error distribution may not be normally distributed ~~and~~ giving rise to non constant variance - covariance metrix discussed by Box[9].

However, since simulated data for estimated value of concentration were available the second alternative was considered for data generation with the rate vector as response vector and also under these circumstances it may be justifiably assumed that the error of observation will follow a multivariate normal distribution.

2.2 Estimation of parameters and variance - covariance matrix of observations

The main problems excluding the inherent difficulties, associated with Box and Hill criterion[15,16] for multiresponse situation are the estimation of parameters and prior knowledge of variance covariance matrix.

Koch[22] has discussed the estimation of variance in univariate case and also of covariance in multivariate case. Rao[23] also discussed the estimation of variance covariance matrix from known data for responses. Hill and Hunter[24] extended Box and Hill criterion for single response systems,

analytically, for unknown variance case but no such method was available for multiresponse case. Box and Draper[3] suggested that covariance matrix may be replaced by cofactors of the elements in the dispersion matrix. But since the problem was toestimate parameters as well as covariance matrix simultaneously, the method adopted by Beuchamp and Cornell[6] for parameter estimation was used in the study. The intermediate covariance matrix generated while estimating parameters was used as the estimate for covariance matrix and the underlying theory is given as follows:

Considering the multiresponse model

$$y^{q}_{rj} = f_{rj}\ (\underline{x}^{q}_{rj}\ ,\ \underline{\theta}_{r}\ ) + \epsilon_{jq} \qquad (2.2.1)$$

If $\underset{(N\times 1)}{\underline{\epsilon}_j} = (\epsilon_{j1}\ \epsilon_{j2}\ \cdots\ \epsilon_{jN}\ )^{T}$ and

$$\underset{(Nu\times 1)}{\underline{\epsilon}} = (\epsilon_1'\ \ \epsilon_2'\ \ \ldots\ \ \epsilon_u')^{T} \qquad (2.2.2)$$

are defined and it is assumed that for each j the n components of ϵ_j are generated from a distribution such that

$$E(\underset{(N\times 1)}{\underline{\epsilon}_j}) = \underset{(N\times 1)}{\underline{0}} \quad \text{and} \quad (\underset{(N\times 1)}{\epsilon_j}\ \underset{(1\times N)}{\epsilon_j^{T}}\ ) = \sigma_{jj}\ \underset{(N\times N)}{\underline{\underline{I}}} \quad 0 < \sigma_{jj} < \infty \qquad (2.2.3)$$

and also that the set of u random vectors $\underline{\epsilon}_j$ follows a multivariate distribution with

$$E(\underset{(N\times 1)}{\underline{\epsilon}_j}\ \underset{(1\times N)}{\underline{\epsilon}_i^{T}}\ ) = \sigma_{ij}\ \underset{(N\times N)}{\underline{\underline{I}}} \quad \text{where } -\infty < \sigma_{ij} < \infty \text{ and } i \neq j \qquad (2.2.4)$$

which implies that correlation between observations for different responses is being considered.

Therefore,

$$E(\underset{(Nux1)}{\underline{\epsilon}}\ \underset{(1xNu)}{\underline{\epsilon}^{T}}) = \underset{(NuxNu)}{\underline{\underline{\Omega}}} = \underset{(u\times u)}{\underline{\underline{\Sigma}}} \otimes \underset{(NxN)}{\underline{I}} \qquad (2.2.5)$$

where $\underline{\underline{\Sigma}} = \{\sigma_{ij}\}$ is a symmetric positive definite matrix and $\otimes$ denotes the Kronecker product.

If $\underset{(Nx1)}{Y_j} = (y_{rj}^{1}\ y_{rj}^{2} \ldots y_{rj}^{N})^{T}$ and $\underline{F}_j(\underline{x},\underline{\theta}) = (f_{rj}(\underline{x}_{rj}^{1},\theta)\ldots$

$$f_{rj}(\underline{x}_{rj}^{N}, \theta))^{T}$$

are represented as shown and also it is assumed that $\underline{\epsilon}$ follows a multivariate normal distribution then equation (2.2.1) can be represented as

$$\underline{Y}_j = \underline{F}_j\ (\underline{x},\underline{\theta}) + \underline{\epsilon}_j \qquad (2.2.6)$$

Further, if $\underset{(Nux1)}{\underline{Y}} = (\underline{Y}_1^{T}, \underline{Y}_2^{T} \ldots \underline{Y}_u^{T})^{T}$ (2.2.7)

and $\underset{(Nux1)}{F} = (F_1(\underline{x},\underline{\theta}) \ldots F_u(\underline{x},\underline{\theta}))^{T}$ (2.2.8)

then eqn. (2.2.1) can be more elegantly described as

$$\underset{(NuX1)}{\underline{Y}} = \underset{(NuX1)}{\underline{F}} + \underset{(NuX1)}{\underline{\epsilon}} \qquad (2.2.9)$$

which essentially resembles a single response model.

With the above assumption the maximum likelihood estimate of parameters can be obtained by minimizing

$$L\ (\underline{\theta}) = (\underline{Y} - \underline{F})^T \underline{\underline{\Omega}}^{-1}\ (\underline{Y} - \underline{F}) \tag{2.2.10}$$

with respect to $\underline{\theta}$, where eqn. (2.2.10) is a function of $\underline{\theta}$ only. The suggested estimate for $\underline{\underline{\Sigma}}$ and hence $\underline{\underline{\Omega}}$ is given as

$$\hat{\sigma}_{ij} = \underset{(1XN)}{\underline{\epsilon}_j^T(o)}\ \underset{(NX1)}{\underline{\epsilon}_i(o)}\ /N$$

$$\text{where } \underset{(NX1)}{\underline{\epsilon}_j(o)} = (\epsilon_{j1}(o)\ \ldots\ \epsilon_{jN}(o))^T$$

$$\text{and } \underline{\epsilon}_{jq}(o) = y_{rj}^{q} - f_{rj}(\underline{x}_{rj}^{q}\ ,\ \underline{\hat{\theta}}_r^{(j)}) \tag{2.2.11}$$

where $\hat{\underline{\theta}}^{(j)}$ is the least square estimate of $\underline{\theta}^{(j)}$ and is the vector of parameters for rth model and jth response and $\underline{\theta}^{(j)}$ is a subset of $\underline{\theta}$. The estimate for $\hat{\underline{\theta}}^{(j)}$ is obtained by non-linear regression of function by least square technique.

$$L_j\ (\underline{\theta}^{(j)}) = (\underline{Y}_j - F_j(\underline{x}_{rj}^{q}, \underline{\theta}_r^{j}))^T(\underline{Y}_j - F_j(\underline{x}_{rj}^{q}, \theta_r^{j})) \tag{2.2.12}$$

Modified Gauss-Newton algorithm was suggested for fitting of non linear regression and is ellaborated as follows

$$\underset{(Nux1)}{\underline{F}} = \underset{(Nux1)}{\underline{F}_o} + \underset{(Nuxm)}{\underline{\underline{F}}_o'}\ \underline{\delta}\ o \tag{2.2.13}$$

$$\text{where } \underset{(Nuxm)}{\underline{\underline{F}}'} = (\underline{\underline{F}}_1'\ \ \underline{\underline{F}}_2'\ \ldots\ \underline{\underline{F}}_u')^T \tag{2.2.14}$$

$$\text{and } \underline{\underline{F}}_j' = \{f_{qk}\}\ \ 1 \leq q \leq N,\ 1 \leq k \leq m \text{ and } f_{qk} = \left[\frac{\partial f_j(\underline{x}_q, \underline{\theta})}{\partial \theta_k}\right] \tag{2.2.15}$$

and $\underline{\delta} = \underline{\theta} - \underline{\theta}_o$ (2.2.16)

So that the least square estimate of $\underline{\delta}$ is

$$\underset{(mx1)}{\hat{\underline{\delta}}} = ((\underset{(mxNu)}{\underline{\underline{F}}_o'})^T \underset{(NuxNu)}{\underline{\underline{\Omega}}^{-1}} \ (\underset{(Nuxm)}{\underline{\underline{F}}_o'}))((\underset{(mxNu)}{\underline{\underline{F}}_o'})^T \ \underset{(NuxNu)}{\underline{\underline{\Omega}}^{-1}} \ \underset{(Nux1)}{\underline{Y}}) \quad (2.2.17)$$

The current estimate of vector $\underline{\theta}_o$

$$c\,\underline{\theta}_o = \underline{\theta}_o + c\,\hat{\underline{\delta}} \text{ where } o \leq c \leq 1 \quad (2.2.18)$$

when substituted in eqn. (2.2.18) for obtaining the next current value of $\underline{\theta}$ given by

$$c\,\underline{\theta}_1 = \underline{\theta}_o + c_{min}\,\hat{\underline{\delta}} \quad (2,2.19)$$

and the iterative process is carried out until $\underline{\theta}$ is approximated with sufficient accuracy. However, as other non linear regression methods, a feasible preliminary estimate of $\underline{\theta}_o$ is necessary for convergence.

In the present study all the models are linear with respect to parameters and method followed by Ray[1] is adopted for estimation of parameters.

After σ_{ij} is obtained which gives the variance covariance matrix of observation between responses, $\underline{\underline{\Omega}}$ is obtained from eqn. (2.2.20)

$$\underset{(nuxnu)}{\underline{\underline{\Omega}}} = \underset{(uxu)}{\underline{\underline{\Sigma}}} \otimes \underset{(nxn)}{\underline{\underline{I}}} \quad (2.2.20)$$

and estimated values of parameters given by

$$\hat{\underline{\theta}} = (\underline{\underline{X}}^T \hat{\underline{\underline{\Omega}}} \ \underline{\underline{X}})^{-1} \ \underline{\underline{X}}^T \ (\hat{\underline{\underline{\Omega}}})^{-1} \ \underline{Y} \qquad (2.2.21)$$

This method enables one to get simultaneous estimates of parameters $\hat{\theta}$ and $\hat{\underline{\underline{\Sigma}}}$ which are essential in applying Box and Hill criterion for model discrimination.

2.3 Sequential Design of experiments

A general multiresponse situation represents the condition in which u multivariate observations or responses are being measured where the responses are functions of some independent variables, intrinsic parameters and, in addition, are also associated with the inherent error of observations.

The above idea can be expressed mathematically in a more elegant way as follows

For any model r,

$$y^q_{rj} = f_{rj} \ (\underline{x}^q_{rj}, \ \underline{\theta}_r) + \epsilon_{jq} \qquad (2.3.1)$$

where y^q_{rj} is the jth response for the qth experimental condition,

$f_{rj}(\underline{x}^q_{rj}, \underline{\theta}_r)$ is the expected value of y_{jq},

$\underline{x}^q_{rj}$ is the vector of n independent variables for the qth experiment,

$\underline{\theta}_r$ is the vector of the intrinsic parameter having m components,

ϵ_{jq} is the random experimental error of observation associated with the jth response and the qth experiment.

The limits for the suffixes are

(i) $1 \leq r \leq v$, where v number of models are considered

(ii) $1 \leq j \leq u$, where u n mber of responses are observed

(iii) $N \leq q \leq N+N^*$, where N set of observations have already been made and design conditions for further N^* set are to be selected.

N=0 implies that some prior information is already available.

Also that

(iv) $\underline{\theta}_r = \{\theta_k\}$, $1 \leq k \leq m$, where the parameters may or may not be same for all the models.

(v) $\underline{x} = \{x_h\}$, $1 \leq h \leq n$

(vi) The random error ϵ_{jq} is assumed to follow multivariate normal distribution, and

$$E(\epsilon_{jq})=0 \text{ for all } j,q$$

$$E(\epsilon_{iq}\ \epsilon_{js}) = \sigma_{ij} \text{ for all } i,j,\ q=s \qquad (2.3.2)$$

$$= 0 \text{ for all } i,j,\ q \neq s$$

The last condition means that any two experiments q,s are considered to be independent of each other i.e., they are uncorrelated observations.

In vector notations, let

$Y^T = (y_1 \; \dots \; y_j \; \dots \; y_u)$ be the vector of observation,

$f_r^T = (f_{r1} \dots f_{rj} \dots f_{ru})$ be the vector of the expected values of observation for the rth model,

$f^T = (f_1^T \dots f_r^T \dots f_v^T)$ be the vector of the expected values of observation for all the models.

It is assumed that the observations Y are normally distributed about their expected values f_r with a variance covariance matrix

$\underline{\underline{\Sigma}}_r = \{\sigma_{ij}\}$, $1 \leq i \leq u$, $1 \leq j \leq u$, then the probability density for Y^{N+1} is

$$p(f^{N+1} \mid f_r, \Sigma_r) = \frac{|\Sigma_r|^{-1/2}}{(2\pi)^{u/2}} \exp\left[-\frac{1}{2}(Y^{N+1} - f_r)^T \Sigma_r^{-1} (Y^{N+1} - f_r)\right] \tag{2.3.3}$$

Now, if f_r is linear in the parameter space $\underline{\theta}$, or can be approximately expressed as a linear function of $\underline{\theta}$ in the region of the parameter estimates $\hat{\underline{\theta}}$, then one gets

$$f_{rj}^N = f_{rj}(\underline{\hat{\theta}}_r^N, \underline{x}_{rj}^N) + \sum_{k=1}^{m} (\theta_{rk}^N - \hat{\theta}_{rk}^N) \left[\frac{\partial f_{rj}(\hat{\theta}_r^N, x_{rj}^N)}{\partial \theta_{rk}}\right]_{\underline{\theta}_r = \underline{\hat{\theta}}_r^N} \tag{2.3.4}$$

Where the $\underline{\hat{\theta}}_r$ are estimated values of $\underline{\theta}$ based on the available N experiments (if N=0, then estimates of $\underline{\theta}$ must be known).

$$f_{rj}^{N} = f_{rj}(\underline{x}_{rj}^{N}, \hat{\underline{\theta}}_{r}^{N}) + \sum_{k=1}^{m} (\theta_{rk} - \hat{\theta}_{rk})^{N} X_{rj,k}^{N} \qquad (2.3.5)$$

where

$$X_{rj,k}^{N} = \left[\frac{\partial f_{rj}(\underline{x}_{rj}^{N}, \hat{\underline{\theta}}_{r}^{N})}{\partial \theta_{rk}} \right]_{\underline{\theta}_r = \hat{\underline{\theta}}_r^{N}} \qquad (2.3.6)$$

and
$$\underline{\underline{X}}_{rj} = \begin{bmatrix} X_{rj,1}^{1} & X_{rj,2}^{1} & \cdots & X_{rj,m}^{1} \\ \vdots & \vdots & & \vdots \\ X_{rj,1}^{N} & X_{rj,2}^{N} & & X_{rj,m}^{N} \end{bmatrix} \qquad (2.3.7)$$

Draper and Hunter[4,5] have shown that the posterior density function for $\underline{\theta}$, after N statistically independent runs, is,

$$p(\underline{\theta} \mid \Sigma_r, Y) = \frac{|M|^{1/2}}{(2\pi)^{m/2}} \exp\left[-\tfrac{1}{2}(\underline{\theta} - \hat{\underline{\theta}})^{T} M (\theta - \hat{\theta})\right] \qquad (2.3.8)$$

where

$$M = \sum_{j=1}^{u} \sum_{l=1}^{u} \sigma^{ij} \underline{\underline{X}}_{rj}^{T} \underline{\underline{X}}_{rl} \qquad (2.3.9)$$

where σ^{ij} is an element of Σ_r^{-1}.

This implies that $(\underline{\theta} - \hat{\underline{\theta}})$ is normally distributed about $\underline{0}$. With covariance matrix M^{-1}.

For convenience, let

$$\underline{\underline{X}}_{r}^{N+1} = \begin{bmatrix} X_{r1,1}^{N+1} & X_{r1,2}^{N+1} & \cdots & X_{r1,m}^{N+1} \\ \vdots & \vdots & & \vdots \\ X_{ru,1}^{N+1} & X_{ru,2}^{N+1} & & X_{ru,m}^{N+1} \end{bmatrix} \qquad (2.3.10)$$

Then it has been proved[16] that $\underline{\underline{X}}_r^{N+1}(\Theta - \hat{\Theta})$ is normally distributed about $\underline{0}$ with a covariance matrix of

$$W_r^{N+1} = (X_r^{N+1})\, M^{-1}\, (X_r^{N+1})^T \qquad (2.3.11)$$

Also, because of the linearization of the models in the parameter space, f_r is normally distributed about the predicted response $\hat{Y}_r^{N+1}$ with a covariance matrix W_r^{N+1} and the probability density function is given by

$$p(f_r \mid \Sigma_r) = \frac{|W_r^{N+1}|^{-1/2}}{(2\pi)^{u/2}} \exp\left[-\frac{1}{2}(f_r - \hat{Y}_r^{N+1})^T (W_r^{N+1})^{-1} (f_r - \hat{Y}_r^{N+1})\right] \qquad (2.3.12)$$

which finally reduces to(16)

$$p(Y_r^{N+1} \mid \Sigma_r) = \frac{|\Sigma_r^{N+1}|^{-1/2}}{(2\pi)^{u}} \exp\left[-\frac{1}{2}(Y^{N+1} - \hat{Y}^{N+1})^T (\Sigma_r^{N+1})^{-1} (Y^{N+1} - \hat{Y}_r^{N+1})\right] \qquad (2.3.13)$$

where

$$\Sigma_r^{N+1} = \Sigma_r + W_r^{N+1} \qquad (2.3.14)$$

The following two relations with equation (2.3.13) gives

$$E\left\{(Y^N - \hat{Y}_r^N)^T (\Sigma_r^N)^{-1} (Y^N - \hat{Y}_r^N)\right\} = \left[E\left\{(Y^N - \hat{Y}_r^N)^T\right\}\right] (\Sigma_r^N)^{-1} \left[E\left\{(Y^N - \hat{Y}_r^N)\right\}\right] = \text{trace } \Sigma_r^N (\Sigma_r^N)^{-1}$$

$$= \text{trace } I_u,$$

I_u is a (uxu) identity matrix

$$E\left\{(Y^N-\hat{Y}_r^N)^T(\Sigma_s^N)^{-1}(Y^N-\hat{Y}_r^N)\right\}=\left[E\left\{(Y^N-\hat{Y}_r^N)^T\right\}\right](\Sigma_s^N)^{-1}\left[E\left\{(Y^N-\hat{Y}_r^N)\right\}\right]$$

$$= \text{trace}\, \Sigma_r^N (\Sigma_s^N)^{-1}$$

In Kullback[21] criterion for discrimination between two hypothesis

$$J^{N+1}(r:s) = I^{N+1}(r:s) + I^{N+1}(s:r) \tag{2.3.15}$$

where $I^{N+1}(r:s) = \int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty} p_r(y) \ln \frac{p_r(y)}{p_s(y)}\, dy^{N+1}$

$$= \frac{1}{2}\left[\ln\left|\frac{\Sigma_s^{N+1}}{\Sigma_r^{N+1}}\right| - \text{trace}\, I_u + \text{trace}\, \Sigma_r^{N+1}(\Sigma_s^{N+1})^{-1} + (\hat{Y}_r^{N+1}-\hat{Y}_s^{N+1})^T(\Sigma_s^{N+1})^{-1}(\hat{Y}_r^{N+1}-\hat{Y}_s^{N+1})\right] \tag{2.3.16}$$

A similar expression is obtained for $I^{N+1}(s:r)$ where the indexes of equation (2.3.16) are only interchanged. Maximizing $J^{N+1}(r:s)$ in eqn. (2.3.15) w.r.t. independent variables one gets the next suitable point for further experimentation. In this approach no weight was given for the prior probabilities and Box and Hill removed the deficiency by maximizing a scalar discriminant function given as

$$K_v = \left[P_1^N\ P_2^N\ \dots\ P_v^N\right]\begin{bmatrix} I(1:1) & I(1:2) & \dots & I(1:v) \\ I(2:1) & I(2:2) & \dots & I(2:v) \\ \vdots & \vdots & & \vdots \\ I(v:1) & I(v:2) & \dots & I(v:v) \end{bmatrix}\begin{bmatrix} P_1^N \\ P_2^N \\ \vdots \\ P_v^N \end{bmatrix}$$

where P denotes the prior probabilities for the models concerned and also that elements on the main diagonal of the I(r:s) matrix is zero.

The expression for K_v reduces to

$$K_v = P_1 P_2\, J(1,2) + P_1 P_3\, J(1,3) + \ldots + P_1 P_v\, J(1,4) + P_2 P_3\, J(2,3) + \ldots + P_{v-1}\, P_v\, J(v-1,v) \qquad (2.3.17)$$

By use of equation (2.3.17) with eqn. (2.3.16) the expression for K_v takes the form

$$K_v = \frac{1}{2} \sum_{r=1}^{v} \sum_{s=r+1}^{v} P_r^N\, P_s^N \left[\text{trace}\left\{ \Sigma_r^{N+1} (\Sigma_s^{N+1})^{-1} + \Sigma_s^{N+1} (\Sigma_r^{N+1})^{-1} - 2I_u \right\} + (\hat{Y}_r^{N+1} - \hat{Y}_s^{N+1})^T \left\{ (\Sigma_s^{N+1})^{-1} + (\Sigma_r^{N+1})^{-1} \right\} (\hat{Y}_r^{N+1} - \hat{Y}_s^{N+1}) \right] \qquad (2.3.18)$$

Eqn. (2.3.18) is used as a design criterion for choosing the independent variables for the (N+1)th run so as to effect discrimination between the rival models.

The steps involved for discriminations among rival models using the discussed method of approach are as follows:

(1) Experiments are carried out for N runs either based on judgement or by some arbitrary or suboptimal way.

(2) Based on these observed values of responses and the models under consideration, the parameters are estimated using linear or non linear regression techniques.

The covariance matrix Σ_r is generally unknown and so appropriate estimates are to be made (discussed in section 2.2).

(3) Values of prior probabilities are to be calculated for (N+1)th run which are the posterior probabilities after the Nth run. To begin with the discrimination process, initial prior probabilities are assumed, based on prior information and otherwise all are taken to be equal to 1/v.

(4) K_v is maximized and the corresponding set of independent variables $\underline{x}^{N+1}$ is taken as the operating condition for the (N+1)th run.

(5) Posterior probabilities after N+1 run are calculated using suitable methods (discussed in section 2.5).

(6) (i) If P_r^{N+1} reaches some value such that it can be accepted by some criterion, experimentation is stopped.

(ii) If the trend of change of P_r^{N+1} is observed to follow some acceptable pattern, experimentation is stopped.

(iii) If P_r^{N+1} does not satisfy (i) or (ii), experiment is conducted at $\underline{x}^{N+1}$ and from step 2 onwards, all steps are are repeated until some model satisfies 6(i) or 6(ii). As the number of experiments goes on increasing one can delete models with very low posterior probabilities and also introduce other plausible models, depending on the judgement of the experimenter.

2.4 Maximization of the scalar discriminant function K_v : Optimization Technique

In the present study the scalar discriminant function K_v is to be maximized w.r.t. the vector of independent variables or controlled variables $\underline{x}$. This essentially enables one to find the design condition $\underline{x}$ for the (N+1)th run and ensures that optimum discrimination takes place between the rival models.

Efforts were made to standard optimization techniques but the following factors were considered before proper selection of the method was made.

(A) Prior information of K_v :

Since K_v is a multivariable function, a knowledge of the nature of the response surface in the co-ordinate space is essential because an initial point, x^0, :? is necessary for starting the search methods. Information of K_v provides a feasible starting point and saves computer time. No such prior information was available which could lead to an erroneous optimum point.

(B) Derivatives of K_v :

The functional dependency of K_v on the vector of independent variables $\underline{x}$, was complicated in nature and no attempts were made to calculate the analytical derivatives of K_v. But attempts to calculate the numerical derivatives

using finite difference methods gave serious problems and the most important of them being the stability and selection of step size. Also, the advantage of the knowledge of numerically calculated derivatives are off-set by the time involved for computation.

(C) <u>Operability region</u>:

Though the maximization of a function gives the feeling of global maxima, in engineering problems, however, these are seldom achieved. This may be attributed to the fact that the values of the independent variables predicted by optimization technique for a global maxima are not feasible in practical cases. For example it is not advisable to conduct a kinetic reaction at a temperature of, say, 2000°C and then to change the design condition to a temperature of -200°C in the next run where both the design conditions are predicted by theoretical considerations.

In fact, while optimizing real problems, the co-ordinate space in which the function is to be maximized is bound by constraints, the values of which depend on the judgement of the experimenter. This space is called the operability region.

Considering the factor (C) the following constraints were imposed on the independent variables

$$
\begin{aligned}
&\text{(i)} \quad 0 \leq x_1 \leq 30 \\
&\text{(ii)} \quad 0 \leq x_2 \leq 30 \qquad\qquad (2.4.1) \\
&\text{(iii)} \quad 0 \leq x_3 \leq 30
\end{aligned}
$$

Where x_1, x_2 and x_3 represent the concentration of A,B and C respectively in gm. moles/litre. These assumptions are justified in the case of kinetic reactions in liquid or gas phase, conducted in a laboratory scale batch reactor.

Study of the function K_v for different values of $\underline{x}$ were made only in the operability region, instead of the whole space. This study helped to choose a feasible starting point. However, factors (B) and (C) restricted the choice to get suitable derivative free multivariable constrained optimization technique.

Complex algorithm by Box[25,26] was used in the present study where the method maximizes a non linear multivariable function subject to non-linear inequality constraints. Mathematically, a function $F(x_1, x_2 \ldots x_n)$ is maximised with respect to n independent variables $x_1, x_2, \ldots x_n$ subject to m constraints of the form

$$g_k \leq x_k \leq h_k \ , \ k = 1, \ldots m \qquad (2.4.2)$$

An initial point $\underline{x}^o$ is to be supplied and thereafter a complex of $k \geq n + 1$ points is formed inclusive of the initial point. The additional (k-1) points are generated using random numbers and the constraints for the independent variables are as

$$x_{ij} = g_i + r_{ij} \, (h_i - g_i) \qquad (2.4.3)$$

where $i = 1, \ldots\ n$

$j = 1, \ldots\ k-1$, represents the vertex

and r_{ij} are random numbers distributed over the interval (0,1).

The points selected in this manner must satisfy the explicit constraints but may not satisfy the implicit constraints. If a point does not satisfy any of the implicit constraints, the point is moved half-way towards the centroid of the remaining points and this modification for the search point is continued until a satisfactory point is reached.

$$x_{ij}(\text{new}) = (x_{ij}\ (\text{old}) + x_{i,c})/2 \quad i=1,\ldots N \qquad (2.4.4)$$

$$\text{where } x_{i,c} = \frac{1}{k-1}\left[\sum_{j=1}^{K} x_{ij} - x_{ij}(\text{old})\right] \quad i=1,\ldots N \qquad (2.4.5)$$

The search technique is to compare the functional values over the constrained region or operability region. Initially the functional values are evaluated at each of the K vartices and the vertex which gives the lowest function value is discarded. The discarded point is replaced by another point $\alpha \geqslant 1$ times as far from the centroid of the remaining points as the reflection of the discarded point in the centroid. The new point thus obtained lines on the line joining the rejected point and the centroid.

$$x_{ij}(\text{New}) = \alpha\,(x_{i,c} - x_{ij}(\text{old})) + x_{ic}, \quad i=1,\ldots n \qquad (2.4.6)$$

However, if the new point repeats itself in giving the least functional value then it is moved halfway towards the centroid to give a new trial point and is repeated until some constraint is violated. If the trial point violates any explicit constraint then the variable is reset to a value $\delta = 0.001$ inside the limit. Choice of α and k depends on some factors which has been extensively dealt with in the orginal work by Box[25] and depends mainly on the experience and the nature of the function subjected to the appropriate constraints. The use of over reflection factor, $\alpha > 1$, tends to cause an enlargement of the complex and thereby compensating for the shrinkage caused by the movement of the complex towards the centroid. Also if the initial point is far removed from the optimum it enhances the progress of the complex towards the optimum. Similarly $k > n+1$ helps the complex from collapsing and with simultaneous use of $\alpha > 1$ helps the complex in maintaining its progress while it reaches some corner.

The stopping criterion or the attainment of maxima was assumed when the function values of each vertex are within β units for γ consecutive operations. Also there was no systematic search for alternative maxima, the maximization was ensured by using more than one satisfactory initial point.

It may be emphasized that complex method by Box provided a good search technique to get the constrained optimization, though not a perfect method.

2.5 Posterior probabilities for the models

The probabilities of all the competing models after the (N+1)th run was updated using Bayes Theorem which is given below

$$P_r^{N+1} = \frac{P_r^{N} L_r}{\sum_{r=1}^{v} P_r^{N} L_r} \qquad (2.5.1)$$

where P_r^{N+1} is the posterior probability and P_r^{N} is the prior probability of the rth model, L_r being the likelihood of the rth model, v is the total number of competing models.

If the values of the posterior probabilities of the models were below the accepted values the method of locating subsequent design condition was carried out via the steps for estimation of parameters and variance covariance matrix of observations until an acceptable value of the posterior probability for a particular model was attained.

CHAPTER 3

RESULTS AND DISCUSSIONS :

3.1 Data Generation.

Nine sets of data were considered to test the Box and Hill's criterion for model discrimination in multiresponse situation.

The scheme adopted for data generation is given below:

(1) Model Used : $A \xrightarrow{\theta_{31}} B \xrightarrow{\theta_{32}} C$

Data Set 1

(i) Parameter values : $\theta_{31} = 0.1\ \text{min}^{-1}$ $\theta_{32} = 0.1\ \text{min}^{-1}$

(ii) $\underline{\underline{C}}$ matrix (eqn.2.1.3): $\begin{pmatrix} .005 & .0 \\ .0 & .003 \end{pmatrix}$

Data Set 2

(i) Parameter values : $\theta_{31} = 0.1\ \text{min}^{-1}$ $\theta_{32} = 0.1\ \text{min}^{-1}$

(ii) $\underline{\underline{C}}$ matrix : $\begin{pmatrix} .003 & .003 \\ .003 & .003 \end{pmatrix}$

Data Set 3

(i) Parameter values : $\theta_{31} = 0.2\ \text{min}^{-1}$ $\theta_{32} = 0.1\ \text{min}^{-1}$

(ii) $\underline{\underline{C}}$ matrix $\begin{pmatrix} .003 & .0 \\ .0 & .003 \end{pmatrix}$

Data Set 4

(i) Parameter values : $\theta_{31} = 0.2\ \text{min}^{-1}$ $\theta_{32} = 0.1\ \text{min}^{-1}$

(ii) $\underline{\underline{C}}$ matrix : $\begin{pmatrix} .003 & .003 \\ .003 & .003 \end{pmatrix}$

(2) Data Set 5

Data reported (19) using model $A \longrightarrow B \longrightarrow C$

(3) Model Used : $A \xrightarrow{\theta_{11}} B \underset{\theta_{13}}{\overset{\theta_{12}}{\rightleftharpoons}} C$

Data Set 6 : $\underline{\underline{C}}$ matrix $\begin{pmatrix} .005 & .0 \\ .0 & .003 \end{pmatrix}$

(i) Parameter values : θ_{11}=0.08 min^{-1} θ_{12}=.02 min^{-1} θ_{13}=.06 min^{-1}

(ii) $\underline{\underline{C}}$ matrix $\begin{pmatrix} .005 & .0 \\ .0 & .005 \end{pmatrix}$

Data Set 7

(i) Parameter values : θ_{11}= .04 min^{-1} θ_{12}=.1 min^{-1} θ_{13}=.2 min^{-1}

(ii) $\underline{\underline{C}}$ matrix : $\begin{pmatrix} .005 & .0 \\ .0 & .005 \end{pmatrix}$

Data Set 8.

(i) Parameter values : θ_{11}= .02 min^{-1} θ_{12}=.02 min^{-1} θ_{13}=.01 min^{-1}

(ii) $\underline{\underline{C}}$ matrix : $\begin{pmatrix} .005 & .005 \\ .005 & .005 \end{pmatrix}$

Data Set 9.

(i) Parameter values : θ_{11}= .03 min^{-1} θ_{12}= .01 min^{-1} θ_{13}=.01 min^{-1}

(ii) $\underline{\underline{C}}$ matrix : $\begin{pmatrix} .005 & .005 \\ .005 & .005 \end{pmatrix}$

During data generation, the main consideration was that the data should maintain the physical behaviour as far as possible. That is corresponding to an increase in concentration of a particular component, the response which is the rate of change of concentration should be positive and vice versa. However in practice this consideration doesnot hold good specially for small changes in concentration and hence for smaller values for rates. Thus to simulate real situation, it was observed that a value of 0.003 to 0.005 for the elements of $\underline{\underline{C}}$ matrix sufficed the requirement. The logic was that the error introduced with the actual rate was of the form $\underline{\underline{C}}\ \underline{e}$, where $\underline{e}$ is the vector of random normal deviates and the largest element in $\underline{e}$ was roundabout

$\pm$ 2.5, and thus the magnitude of largest error that can occur was nearly $\pm$ .008 to $\pm$ .013. It should be mentioned that in the present study the response generally varied from .8 to .001 and thereby the higher rates were less affected whereas the lower rates were affected increasingly, justifying the assumptions made.

Initially 15 runs were generated using the scheme as already explained and are shown in Tables 3.1 through Table 3.9 where x_1, x_2, x_3 represent the concentration of A,B and C respectively in gm moles/litre and Rate 2, Rate 3 represent the responses of B and C respectively in gm moles/litre min. The models given be equation 2.1 were considered as the competing models and the parameters and variance covariance matrix of observation were next estimated.

3.2 Estimation of Parameters and Variance -Covariance matrix.

The method used for the estimation purpose was developed by Beauchamp and Cornell[6]. The computer program used for the above purpose was developed by Ray[1] and proper modifications were made in it to suit the present study. The estimates for θ and Σr are given in Tables 3.10 through 3.15.

As seen from Table 3.12 corresponding to model 3, data set 1 and run 15, the estimated values of the parameters are 0.1004 min^{-1} and .0998 min^{-1} which are very close to the actual values of the parameters .1 min^{-1} and .1 min^{-1}, used for data generation. But for the estimates of $\hat{\Sigma} r$, the $\hat{\Sigma} r_{22}$ element i.e. the 4th element in the column for $\hat{\Sigma} r$ is seen

Table 3.1

DATA SET 1

Model $A \xrightarrow{\theta_{31}} B \xrightarrow{\theta_{32}} C$ $\theta_{31} = .1 \text{ min}^{-1}$ $\theta_{32} = .1 \text{ min}^{-1}$ $\underline{C} = \begin{pmatrix} .005 & 0 \\ 0 & .003 \end{pmatrix}$

RUN No.	x_1*	x_2	x_3	RATE 2*	RATE 3
1	6.703	2.681	0.615	0.405	0.271
2	4.493	3.595	1.912	0.081	0.357
3	3.012	3.614	3.374	-0.056	0.366
4	2.019	3.230	4.751	-0.118	0.326
5	1.353	2.707	5.940	-0.134	0.272
6	0.907	2.177	6.916	-0.128	0.217
7	0.608	1.703	7.689	-0.113	0.166
8	0.408	1.304	8.743	-0.089	0.131
9	0.273	0.984	8.288	-0.071	0.098
10	0.183	0.733	8.743	-0.063	0.066
11	0.123	0.540	9.084	-0.036	0.059
12	0.082	0.395	9.337	-0.031	0.040
13	0.055	0.287	9.522	-0.019	0.033
14	0.037	0.207	9.756	-0.015	0.023
15	0.025	0.149	9.826	-0.016	0.011
16	0.001	29.999	0.001	-2.991	2.995
17	0.000	29.999	3.099	-3.006	3.011
18	0.001	29.999	18.579	-2.998	3.005
19	0.001	29.999	18.579	-3.006	3.012
20	9.222	0.001	29.999	0.936	0.004

* x are in gm moles/l .

Rates are in gm mole/l..min.

Table 3.2

DATA SET 2

Model : $A \xrightarrow{\theta_{31}} B \xrightarrow{\theta_{32}} C$ $\theta_{31} = .1 \ \text{min}^{-1}$ $\theta_{32} = .1 \ \text{min}^{-1}$ $\underline{\underline{C}} = \begin{pmatrix} .003 & .003 \\ .003 & .003 \end{pmatrix}$

RUN No.	x_1	x_2	x_3	Rate 2	Rate 3
1	6.703	2.681	0.615	0.406	0.272
2	4.493	3.359	1.912	0.090	0.359
3	3.011	3.614	3.373	-0.055	0.366
4	2.019	3.230	4.750	-0.123	0.320
5	1.353	2.706	5.939	-0.130	0.275
6	0.907	2.177	6.915	-0.125	0.219
7	0.608	1.702	7.689	-0.108	0.171
8	0.407	1.304	8.288	-0.090	0.129
9	0.273	0.983	8.743	-0.070	0.098
10	0.183	0.732	9.084	-0.060	0.067
11	0.122	0.540	9.337	-0.035	0.060
12	0.082	0.395	9.522	-0.028	0.041
13	0.055	0.286	9.658	-0.018	0.033
14	0.037	0.207	9.755	-0.015	0.022
15	0.024	0.148	9.826	-0.017	0.010
16	0.001	29.999	3.799	-3.031	3.035
17	0.001	29.999	17.382	-3.060	3.065
18	0.001	29.999	29.637	-3.045	3.051
19	0.001	29.999	0.001	-3.060	3.066
20	25.713	0.001	29.999	2.690	0.004

Table 3.3

DATA SET 3

Model : $A \xrightarrow{\theta_{31}} B \xrightarrow{\theta_{32}} C$ $\quad \theta_{31} = .2 \text{ min}^{-1}$ $\theta_{32} = .1 \text{ min}^{-1}$ $\underline{\underline{C}} = \begin{pmatrix} .003 & 0 \\ 0 & .003 \end{pmatrix}$

Run No.	x_1	x_2	x_3	Rate 2	Rate 3
1	4.493	4.420	1.087	0.459	0.444
2	2.019	4.949	3.032	-0.093	0.493
3	0.907	4.209	4.883	-0.235	0.425
4	0.408	3.223	6.370	-0.238	0.325
5	0.183	2.340	7.476	-0.196	0.235
6	0.082	1.105	8.208	-0.150	0.164
7	0.037	1.142	8.821	-0.111	0.110
8	0.017	0.782	9.201	-0.075	0.078
9	0.007	0.531	9.461	-0.051	0.053
10	0.003	0.360	9.637	-0.043	0.028
11	0.001	0.242	9.756	-0.019	0.030
12	0.001	0.163	9.836	-0.016	0.017
13	0.001	0.110	9.890	-0.007	0.015
14	0.001	0.074	9.926	-0.005	0.009
15	0.001	0.049	9.950	-0.009	0.001
16	0.001	29.999	0.001	-3.021	3.026
17	0.001	29.999	29.999	-3.046	3.051
18	0.001	29.999	3.470	-3.033	3.039
19	0.001	29.999	29.999	-3.046	3.053
20	29.999	7.132	29.999	-5.592	0.726

Table 3.4

DATA SET 4

Model : $A \xrightarrow{\theta_{31}} B \xrightarrow{\theta_{32}} C$ θ_{31} = .2 min^{-1} θ_{32} = .1 min^{-1} $\underline{\underline{C}} = \begin{pmatrix} .003 & .003 \\ .003 & .003 \end{pmatrix}$

Run No.	x_1	x_2	x_3	Rate	Rate
1	4.493	4.419	1.086	0.460	0.446
2	2.019	4.948	3.032	-0.091	0.495
3	0.907	4.209	4.883	-0.234	0.425
4	0.407	3.222	6.369	-0.243	0.319
5	0.183	2.340	7.476	-0.192	0.238
6	0.082	1.649	8.267	-0.146	0.166
7	0.037	1.142	8.820	-0.105	0.115
8	0.016	0.782	9.201	-0.075	0.077
9	0.007	0.531	9.461	-0.051	0.053
10	0.003	0.359	9.637	-0.041	0.030
11	0.001	0.242	9.756	-0.017	0.030
12	0.000	0.163	9.836	-0.013	0.018
13	0.000	0.109	9.890	-0.006	0.015
14	0.000	0.073	9.926	-0.005	0.009
15	0.000	0.049	9.950	-0.009	0.000
16	29.999	0.001	29.999	6.006	3.049
17	0.001	29.999	10.577	-3.004	3.009
18	0.001	29.999	13.073	-3.004	3.010

Table 3.5

(19) DATA SET 5

Model : A B C

Run No.	x_1	x_2	x_3	Rate 2	Rate 3
1	3.862	0.137	0.000	1.372	0.008
2	3.726	0.271	0.003	1.337	0.018
3	3.464	0.525	0.009	1.276	0.034
4	3.229	0.744	0.026	1.093	0.082
5	3.011	0.941	0.047	0.983	0.108
6	2.807	1.118	0.074	0.884	0.134
7	2.617	1.277	0.105	0.793	0.156
8	2.440	1.419	0.140	0.710	0.175
9	2.121	1.557	0.220	0.596	0.200
10	1.843	1.843	0.311	0.466	0.228
11	1.603	1.985	0.411	0.358	0.249
12	1.393	2.089	0.517	0.259	0.265
13	1.129	2.810	0.690	0.152	0.288
14	0.915	2.208	0.876	0.046	0.311
15	0.740	2.211	1.048	0.005	0.286
16	0.001	29.999	29.999	-3.558	3.563
17	0.009	3.113	29.308	-3.849	3.933

Table 3.6

DATA SET 6

Model : $A \xrightarrow{\theta_{11}} B \underset{\theta_{13}}{\overset{\theta_{12}}{\rightleftharpoons}} C$ $\theta_{11} = .08\ \mathrm{min}^{-1}$ $\theta_{12} = .02\ \mathrm{min}^{-1}$ $\theta_{13} = .06\ \mathrm{mil}^{-1}$ $\underline{\underline{C}} = \begin{pmatrix} .005 & .0 \\ .0 & .005 \end{pmatrix}$

Run No.	x_1	x_2	x_3	Rate 2	Rate 3
1	10.892	5.295	1.813	0.875	- 0.002
2	7.909	8.174	1.916	0.579	0.043
3	5.743	10.092	2.164	0.385	0.070
4	4.171	11.359	2.470	0.256	0.081
5	3.028	12.188	2.784	0.161	0.073
6	2.199	12.723	3.078	0.106	0.070
7	1.597	13.064	3.340	0.064	0.058
8	1.160	13.276	3.565	0.045	0.056
9	0.842	13.405	3.753	0.027	0.045
10	0.611	13.480	3.909	0.006	0.027
11	0.444	13.521	4.035	0.013	0.034
12	0.322	13.541	4.137	0.004	0.023
13	0.234	13.548	4.218	0.004	0.022
14	0.170	13.549	4.281	0.002	0.017
15	0.123	13.545	4.331	0.004	0.017
16	29.999	0.000	29.999	4.315	-1.913
17	0.001	29.999	29.999	1.210	-1.205

Table 3.7

DATA SET 7

Model : $A \xrightarrow{\theta_{11}} B \underset{\theta_{13}}{\overset{\theta_{12}}{\rightleftharpoons}} C$

$\theta_{11} = .04 \text{ min}^{-1}$

$\theta_{12} = .1 \text{ min}^{-1}$

$\theta_{13} = .2 \text{ min}^{-1}$

$\underline{\underline{C}} = \begin{pmatrix} .005 & 0 \\ 0 & .005 \end{pmatrix}$

Run No.	x_1	x_2	x_3	Rate 2	Rate 3
1	12.782	3.601	1.617	0.475	0.037
2	10.892	5.137	1.971	0.310	0.114
3	9.282	6.240	2.478	0.240	0.125
4	7.909	7.118	2.972	0.200	0.119
5	6.740	7.848	3.412	0.163	0.098
6	5.743	8.464	3.792	0.142	0.088
7	4.894	8.988	4.118	0.117	0.072
8	4.171	9.433	4.396	0.107	0.068
9	3.554	9.813	4.633	0.089	0.057
10	3.028	10.136	4.835	0.067	0.038
11	2.581	10.412	5.007	0.069	0.045
12	2.199	10.647	5.154	0.054	0.034
13	1.874	10.847	5.279	0.050	0.032
14	1.597	11.017	5.386	0.042	0.027
15	1.361	11.163	5.477	0.039	0.026
16	29.979	0.000	30.000	7.311	-6.110
17	0.001	29.999	0.001	-3.011	3.016

Table 3.8

DATA SET 8

Model : $A \xrightarrow{\theta_{11}} B \underset{\theta_{13}}{\overset{\theta_{12}}{\rightleftharpoons}} C$ $\quad \theta_{11} = .02 \text{ min}^{-1}$ $\quad \underset{\approx}{C} = \begin{pmatrix} .005 & .005 \\ .005 & .005 \end{pmatrix}$

$\theta_{12} = .2 \text{ min}^{-1}$

$\theta_{13} = .01 \text{ min}^{-1}$

Run No.	x_1	x_2	x_3	Rate 2	Rate 3
1	7.686	0.346	1.967	0.171	-0.008
2	7.095	0.962	1.942	0.145	0.002
3	6.550	1.487	1.963	0.121	0.010
4	6.046	1.933	2.021	0.105	0.021
5	5.581	2.310	2.108	0.084	0.023
6	5.152	2.628	2.220	0.072	0.030
7	4.756	2.894	2.350	0.062	0.036
8	4.390	3.116	2.493	0.052	0.039
9	4.053	3.300	2.648	0.042	0.040
10	3.741	3.450	2.809	0.034	0.041
11	3.454	3.572	2.974	0.029	0.043
12	3.188	3.670	3.142	0.023	0.043
13	2.943	3.748	3.309	0.018	0.043
14	2.717	3.807	3.476	0.012	0.040
15	2.508	3.852	3.640	0.005	0.037
16	0.001	29.999	27.757	-0.330	0.335
17	29.999	0.001	29.999	0.880	-0.257

Table 3.9

DATA SET 9

Model : $A \xrightarrow{\theta_{11}} B \underset{\theta_{13}}{\overset{\theta_{12}}{\rightleftharpoons}} C$

θ_{11} = .03 min^{-1}
θ_{12} = .01 min^{-1}
θ_{13} = .01 min^{-1}

$\underline{C} = \begin{pmatrix} .005 & .005 \\ .005 & .005 \end{pmatrix}$

Run No.	x_1	x_2	x_3	Rate 2	Rate 3
1	9.418	2.537	0.045	0.262	0.029
2	8.353	3.485	0.162	0.220	0.036
3	7.408	4.283	0.308	0.183	0.040
4	6.570	4.952	0.478	0.155	0.047
5	5.827	5.508	0.665	0.124	0.046
6	5.169	5.968	0.864	0.103	0.050
7	4.584	6.344	1.072	0.086	0.054
8	4.066	6.650	1.285	0.070	0.056
9	3.606	6.894	1.500	0.055	0.055
10	3.198	7.086	1.716	0.042	0.053
11	2.837	7.234	1.929	0.034	0.055
12	2.516	7.345	2.139	0.024	0.053
13	2.231	7.423	2.345	0.017	0.052
14	1.979	7.476	2.549	0.009	0.048
15	1.755	7.505	2.739	0.001	0.044
16	17.164	11.897	23.085	0.649	-0.123
17	0.001	29.999	29.999	0.032	-0.027

Table 3.10

Model	Run No.	Data Set 1 $\hat{\theta}$	Data Set 1 r	Data Set 2 $\hat{\theta}$	Data Set 2 $\hat{\Sigma}_r$	Data Set 3 $\hat{\theta}$	Data Set 3 $\hat{\Sigma}_r$	Data Set 4 $\hat{\theta}$	Data Set 4 $\hat{\Sigma}_r$
	15	0.1005	0.1725×10^{-4}	0.1019	0.4644×10^{-4}	0.2010	0.2060×10^{-3}	0.2004	0.1201×10^{-4}
	15	0.0999	0.1725×10^{-4}	0.1021	0.4643×10^{-4}	0.1001	0.2060×10^{-3}	0.0996	0.1201×10^{-4}
		0.0001	0.1725×10^{-6}	0.0004	0.4643×10^{-4}	-0.0003	0.2060×10^{-3}	0.0002	0.1201×10^{-4}
			0.1389×10^{-1}		0.1399×10^{-1}		0.2326×10^{-1}		0.2331×10^{-1}
		0.0999	0.1633×10^{-4}	.1004	$.4394\times10^{-4}$	.2012	$.1934\times10^{-3}$	.2010	$.1127\times10^{-4}$
1	16	0.1003	0.1634×10^{-4}	.1029	$.4394\times10^{-4}$	.1004	$.1934\times10^{-3}$	.1003	$.1131\times10^{-4}$
		0.0000	0.1634×10^{-4}	.0004	$.4394\times10^{-4}$	-.0009	$.1934\times10^{-3}$	.0001	$.1131\times10^{-4}$
			0.4634×10^{-1}		$.4679\times10^{-1}$		$.6049\times10^{-1}$		$.1198\times10^{-1}$
		.1006	$.2175\times10^{-4}$	.1018	$.7138\times10^{-4}$	.2020	$.1860\times10^{-3}$	.2000	$.1201\times10^{-4}$
	17	.1000	$.2175\times10^{-4}$	.1016	$.7137\times10^{-4}$	.1009	$.1861\times10^{-3}$	.1003	$.1200\times10^{-4}$
		.0000	$.2175\times10^{-4}$	.0	$.7138\times10^{-4}$	-.0005	$.1861\times10^{-3}$	.0002	$.1200\times10^{-4}$
			$.4477\times10^{-4}$		$.4632\times10^{-1}$		$.5923\times10^{-1}$		$.1131\times10^{-1}$
		.0999	$.2056\times10^{-4}$	.1000	$.6745\times10^{-4}$	.2016	$.1770\times10^{-3}$	.2009	$.1143\times10^{-4}$
	18	.1003	$.2057\times10^{-4}$	.1023	$.6745\times10^{-4}$	.1007	$.1770\times10^{-3}$	.1003	$.1148\times10^{-4}$
		.0000	$.2057\times10^{-4}$	.0003	$.6745\times10^{-4}$	-.0009	$.1770\times10^{-3}$	.0001	$.1148\times10^{-4}$
			$.4345\times10^{-1}$		$.4464\times10^{-1}$		$.5642\times10^{-1}$		$.1069\times10^{-1}$
		.1012	$.2153\times10^{-4}$	.1043	$.7055\times10^{-4}$	.2126	$.1681\times10^{-3}$		
	19	.1001	$.2153\times10^{-4}$	.1017	$.7055\times10^{-4}$	.1010	$.1681\times10^{-3}$		
		.0	$.2153\times10^{-4}$	.0001	$.7055\times10^{-4}$	-.0005	$.1681\times10^{-3}$		
			$.4167\times10^{-1}$		$.4233\times10^{-1}$		$.5377\times10^{-1}$		

Table 3.11

MODEL	RUN NO	Data Set 1 $\hat{\theta}$	Data Set 1 $\hat{\Sigma}_r$	Data Set 2 $\hat{\theta}$	Data Set 2 $\hat{\Sigma}_r$	Data Set 3 $\hat{\theta}$	Data Set 3 $\hat{\Sigma}_r$	Data Set 4 $\hat{\theta}$	Data Set 4 $\hat{\Sigma}_r$
		0.1003	0.1841×10^{-3}	0.0998	0.4421×10^{-3}	0.2016	0.1647×10^{-1}	0.1989	0.1841×10^{-3}
	15	-0.0003	0.1727×10^{-4}	-0.0040	0.4789×10^{-4}	0.0002	0.2155×10^{-3}	-0.0016	0.1284×10^{-4}
		0.0999	0.1727×10^{-4}	0.1022	0.4789×10^{-4}	0.1011	0.2155×10^{-3}	0.0997	0.1284×10^{-4}
			0.1688×10^{-1}		0.1689×10^{-1}		0.2678×10^{-1}		0.2658×10^{-1}
		0.0997	0.3847×10^{-1}	.0999	$.3460\times10^{-1}$	.2013	$.3630\times10^{-1}$	.2011	$.1817\times10^{-3}$
	16	-0.0002	0.1635×10^{-4}	-.0002	$.4766\times10^{-4}$	-.0001	$.2031\times10^{-3}$	.0003	$.1225\times10^{-4}$
		0.0999	0.1635×10^{-4}	.1012	$.4766\times10^{-4}$	.1009	$.2031\times10^{-3}$	.0001	$.1225\times10^{-4}$
2			0.4849×10^{-1}		$.4885\times10^{-1}$		$.6101\times10^{-1}$		$.2279\times10^{-1}$
		.0998	$.5953\times10^{-1}$	.1002	$.6050\times10^{-1}$	.2019	$.6324\times10^{-1}$	.2002	$.4211\times10^{0}$
	17	-.0001	$.2173\times10^{-4}$	-.0001	$.7153\times10^{-4}$	-.0001	$.2087\times10^{-3}$	-.0011	$.1205\times10^{-4}$
		.1001	$.2173\times10^{-4}$	.1016	$.7153\times10^{-4}$	.1013	$.2087\times10^{-3}$	.1003	$.1205\times10^{-4}$
			$.4476\times10^{-1}$		$.4733\times10^{-1}$		$.5924\times10^{-1}$		$.1131\times10^{-1}$
		.1008	$.3455\times10^{1}$	.1037	$.3381\times10^{1}$	.2068	$.3424\times10^{1}$	.2005	$.6478\times10^{-1}$
	18	-.0002	$.2037\times10^{-4}$	-.0002	$.6747\times10^{-4}$	-.0002	$.1970\times10^{-3}$	-.0002	$.1277\times10^{-4}$
		.1000	$.2037\times10^{-4}$	.1014	$.6747\times10^{-4}$	.1011	$.1970\times10^{-3}$	.1003	$.1277\times10^{-4}$
			$.4457\times10^{-1}$		$.4504\times10^{-1}$		$.5643\times10^{-1}$		$.2059\times10^{1}$
		.0999	.1203	.1002	.1053	.2020	.1096		
	19	-.0001	$.2179\times10^{-4}$	.0	$.7300\times10^{-4}$	-.0001	$.1929\times10^{-3}$		
		.1002	$.2179\times10^{-4}$	.1017	$.7300\times10^{-4}$	.1014	$.1929\times10^{-3}$		
			$.4243\times10^{-1}$		$.4291\times10^{-1}$		$.5378\times10^{-1}$		

Table 3.12

Model	Run No.	Data Set 1 $\hat{\Theta}$	Data Set 1 $\hat{\Sigma}_r$	Data Set 2 $\hat{\Theta}$	Data Set 2 $\hat{\Sigma}_r$	Data Set 3 $\hat{\Theta}$	Data Set 3 $\hat{\Sigma}_r$	Data Set 4 $\hat{\Theta}$	Data Set 4 $\hat{\Sigma}_r$
		0.1004	0.1727×10^{-4}	0.1016	0.4790×10^{-4}	0.2016	0.2155×10^{-3}	0.2001	0.1283×10^{-4}
		0.0998	0.1727×10^{-4}	0.1011	0.4789×10^{-4}	0.1008	0.2155×10^{-3}	0.0993	0.1283×10^{-4}
	15		0.1727×10^{-4}		0.4789×10^{-4}		0.2155×10^{-3}		0.1283×10^{-4}
			0.1688×10^{-1}		0.1689×10^{-1}		0.2678×10^{-1}		0.2658×10^{-1}
		0.0999	0.1636×10^{-4}	0.1001	0.4767×10^{-4}	0.2022	0.2031×10^{-3}	0.2008	0.1212×10^{-4}
		0.1003	0.1636×10^{-4}	0.1021	0.4767×10^{-4}	0.1016	0.2031×10^{-3}	0.1002	0.1220×10^{-4}
3	16		0.1636×10^{-4}		0.4767×10^{-4}		0.2031×10^{-3}		0.1220×10^{-4}
			0.4849×10^{-1}		0.4885×10^{-1}		0.6102×10^{-1}		0.2279×10^{1}
		0.1006	0.2179×10^{-4}	0.1019	0.7154×10^{-4}	0.2021	0.2087×10^{-3}	0.2002	0.1335×10^{-4}
		0.1000	0.2179×10^{-4}	0.1016	0.7154×10^{-4}	0.1012	0.2087×10^{-3}	0.1002	0.1344×10^{-4}
	17		0.2179×10^{-4}		0.7154×10^{-4}		0.2087×10^{-3}		0.1342×10^{-4}
			0.4684×10^{-1}		0.4734×10^{-1}		0.5924×10^{-1}		0.2179×10^{1}
		0.0999	0.2058×10^{-4}	0.1001	0.6760×10^{-4}	0.2022	0.1972×10^{-3}	0.2009	0.1268×10^{-4}
		0.1003	0.2059×10^{-4}	0.1021	0.6760×10^{-4}	0.1016	0.1972×10^{-3}	0.1002	0.1276×10^{-4}
	18		0.2059×10^{-4}		0.6760×10^{-4}		0.1972×10^{-3}		0.1276×10^{-4}
			0.4457×10^{-1}		0.4504×10^{-1}		0.5644×10^{-1}		0.2059×10^{1}
		0.1012	0.2183×10^{-4}	0.1045	0.7302×10^{-4}	0.2104	0.1930×10^{-3}		
		0.1001	0.2183×10^{-4}	0.1017	0.7302×10^{-4}	0.1013	0.1930×10^{-3}		
	19		0.2183×10^{-4}		0.7302×10^{-4}		0.1930×10^{-3}		
			0.4243×10^{-1}		0.4292×10^{-1}		0.5379×10^{-1}		

Table 3.13

Model	Run No.	Data Set 1 $\hat{\theta}$	Data Set 1 $\hat{\Sigma}_r$	Data Set 2 $\hat{\theta}$	Data Set 2 $\hat{\Sigma}_r$	Data Set 3 $\hat{\theta}$	Data Set 3 $\hat{\Sigma}_r$	Data Set 4 $\hat{\theta}$	Data Set 4 $\hat{\Sigma}_r$
		0.1004	0.1720×10^{-2}	0.1003	0.6608×10^{-3}	0.1988	0.4721×10^{-3}	0.1994	0.3361×10^{-2}
	15	-0.0001	0.1725×10^{-4}	-0.0034	0.4643×10^{-4}	-0.0028	0.2060×10^{-3}	-0.0013	0.1201×10^{-4}
		0.1001	0.1725×10^{-4}	0.1028	0.4643×10^{-4}	0.1009	0.2060×10^{-3}	0.1001	0.1201×10^{-4}
		0.0001	0.1389×10^{-1}	0.0003	0.1399×10^{-1}	-0.0004	0.2326×10^{-1}	0.0001	0.2331×10^{-1}
		0.0997	0.3666×10^{-1}	0.0997	0.2273×10^{-1}	0.2015	0.3407×10^{-1}	0.2014	0.1418×10^{-3}
	16	-0.0001	0.1632×10^{-4}	-0.0002	0.4394×10^{-4}	-0.0001	0.1934×10^{-3}	0.0004	0.1136×10^{-4}
4		0.0999	0.1632×10^{-4}	0.1013	0.4394×10^{-4}	0.1008	0.1934×10^{-3}	0.1002	0.1136×10^{-4}
		-0.0000	0.4634×10^{-1}	0.0002	0.4697×10^{-1}	-0.0005	0.6050×10^{-1}	0.0001	0.1199×10^{1}
		0.0999	0.5953×10^{-1}	0.1012	0.8096×10^{-2}	0.2023	0.6114×10^{1}	0.2203	0.4211×10^{0}
	17	-0.0001	0.2173×10^{-4}	-0.0009	0.7138×10^{-4}	0.0003	0.1860×10^{-3}	-0.0008	0.1203×10^{-4}
		0.1001	0.2173×10^{-4}	0.1021	0.7138×10^{-4}	0.1008	0.1860×10^{-3}	0.0916	0.1203×10^{-4}
		-0.0000	0.4477×10^{-1}	0.0002	0.4633×10^{-1}	-0.0004	0.5923×10^{-1}	-0.0205	0.1131×10^{1}
		0.1011	0.2625×10^{1}	0.1041	0.5275×10^{1}	0.2061	0.2203×10^{1}	0.2008	0.5131×10^{-2}
	18	-0.0002	0.2038×10^{-4}	-0.0002	0.6735×10^{-4}	-0.0002	0.1769×10^{-3}	-0.0001	0.1151×10^{-4}
		0.1000	0.2038×10^{-4}	0.1013	0.6735×10^{-4}	0.1011	0.1769×10^{-3}	0.1001	0.1151×10^{-4}
		-0.0001	0.4345×10^{-1}	-0.0002	0.4465×10^{-1}	-0.0000	0.5643×10^{-1}	-0.0004	0.1069×10^{1}
		0.1000	0.3970×10^{0}	0.1001	0.1852×10^{1}	0.2018	0.8551×10^{1}		
	19	0.0000	0.2146×10^{-4}	0.0001	0.7059×10^{-4}	-0.0001	0.1678×10^{-3}		
		0.1001	0.2146×10^{-4}	0.1018	0.7059×10^{-4}	0.1011	0.1678×10^{-3}		
		-0.0002	0.4167×10^{-1}	0.0001	0.4233×10^{-1}	-0.0005	0.5377×10^{-1}		

TABLE 3.14

6

Model	Run No.	Data Set 5 $\hat{\theta}$	Data Set 5 $\hat{\Sigma}_r$	Data Set 6 $\hat{\theta}$	Data Set 6 $\hat{\Sigma}_r$	Data Set 7 $\hat{\theta}$	Data Set 7 $\hat{\Sigma}_r$	Data Set 8 $\hat{\theta}$	Data Set 8 $\hat{\Sigma}_r$	Data Set 9 $\hat{\theta}$	Data Set 9 $\hat{\Sigma}_r$
		0.3689	0.7555×10^{-3}	0.0798	0.1081×10^{-4}	0.0398	0.1088×10^{-4}	0.0203	0.3202×10^{-5}	0.0303	0.2837×10^{-5}
	15	0.1106	0.7555×10^{-3}	0.0210	0.1082×10^{-4}	0.1016	0.1096×10^{-4}	0.0201	0.3202×10^{-5}	0.0103	0.2839×10^{-5}
		-0.0301	0.7555×10^{-3}	0.0639	0.1082×10^{-4}	0.2038	0.1096×10^{-4}	0.0097	0.3202×10^{-5}	0.0108	0.2839×10^{-5}
1			0.3391		0.7149×10^{-1}		0.1150×10^{-1}		0.1404×10^{-2}		0.3121×10^{-2}
		0.3652	0.7293×10^{-3}	0.0799	0.1014×10^{-4}	0.0399	0.1078×10^{-4}	0.0206	0.3053×10^{-5}	0.0305	0.2723×10^{-5}
	16	0.1238	0.7294×10^{-3}	0.0201	0.1010×10^{-4}	0.1005	0.1077×10^{-4}	0.0192	0.3051×10^{-5}	0.0105	0.2725×10^{-5}
		-0.0039	0.7294×10^{-3}	0.0604	0.1010×10^{-4}	0.2011	0.1077×10^{-4}	0.0087	0.3051×10^{-5}	0.0116	0.2725×10^{-5}
			0.4140		0.8396×10^{-1}		0.3485×10^{-1}		0.7298×10^{-2}		0.1133×10^{-1}
		0.3658	0.3454×10^{-1}	0.0787	0.1931×10^{-2}	0.0318	0.1522×10^{-1}	0.0219	0.1108×10^{-1}	0.0290	0.1887×10^{-2}
	15	-0.0194	0.7900×10^{-3}	-0.0006	0.6868×10^{-3}	-0.0040	0.8238×10^{-3}	0.0023	0.3202×10^{-5}	-0.0008	0.1221×10^{-4}
		0.1275	0.7900×10^{-3}	0.0035	0.6868×10^{-3}	0.0065	0.8238×10^{-3}	0.0112	0.3202×10^{-5}	0.0078	0.1221×10^{-4}
			0.5193		0.7669×10^{-1}		0.2205×10^{-1}		0.1404×10^{-2}		0.9682×10^{-2}
2		0.3641	0.3244×10^{-2}	0.0878	0.4674×10^{-1}	0.1231	0.6761	0.0213	0.1326×10^{-2}	0.0367	0.2458×10^{-1}
	16	-0.0058	0.7683×10^{-3}	0.0016	0.4464×10^{-1}	0.0424	0.6741	0.0004	0.3301×10^{-4}	0.0050	0.1938×10^{-2}
		0.1267	0.7683×10^{-3}	0.0038	0.4464×10^{-1}	0.0161	0.6741	0.0112	0.3301×10^{-4}	0.0038	0.1938×10^{-2}
			0.7985		0.1236×10^{-1}		0.3361		0.9124×10^{-2}		0.2239×10^{-1}

TABLE 3.15

Model	Run No.	Data Set 5 $\hat{\theta}$	Data Set 5 Σ_r	Data Set 6 $\hat{\theta}$	Data Set 6 $\hat{\Sigma}_r$	Data Set 7 $\hat{\theta}$	Data Set 7 $\hat{\Sigma}_r$	Data Set 8 $\hat{\theta}$	Data Set 8 $\hat{\Sigma}_r$	Data Set 9 $\hat{\theta}$	Data Set 9 $\hat{\Sigma}_r$
		0.3702	0.7900×10^{-3}	0.0791	0.6868×10^{-3}	0.0354	0.8238×10^{-3}	0.0213	0.1421×10^{-4}	0.0295	0.1221×10^{-4}
	15	0.1187	0.7900×10^{-3}	0.0032	0.6868×10^{-3}	0.0052	0.8238×10^{-3}	0.0121	0.1421×10^{-4}	0.0075	0.1221×10^{-4}
			0.7900×10^{-3}		0.6868×10^{-3}		0.8238×10^{-3}		0.1421×10^{-4}		0.1221×10^{-4}
			0.5193		0.7669×10^{-1}		0.2205×10^{-1}		0.4812×10^{-2}		0.9682×10^{-2}
3		0.3655	0.7683×10^{-3}	0.0864	0.4464×10^{-1}	0.1073	0.6741	0.0216	0.3301×10^{-4}	0.0298	0.1938×10^{-2}
	16	0.1254	0.7683×10^{-3}	0.0044	0.4464×10^{-1}	0.0333	0.6741	0.0121	0.3301×10^{-4}	0.0070	0.1938×10^{-2}
			0.7683×10^{-3}		0.4464×10^{-1}		0.6741		0.3301×10^{-4}		0.1938×10^{-2}
			0.7985		0.1236×10^{1}		0.3361×10^{1}		0.9124×10^{-2}		0.2239×10^{-1}
		0.3629	0.6756×10^{-1}	0.0798	0.1480×10^{-2}	0.0396	0.2531×10^{-2}	0.0206	0.1108×10^{-1}	0.0306	0.7050×10^{-2}
	15	-0.0197	0.7555×10^{-3}	-0.0001	0.1082×10^{-4}	-0.0003	0.1096×10^{-4}	0.0007	0.3202×10^{-5}	0.0002	0.2839×10^{-5}
		0.1142	0.7555×10^{-3}	0.0210	0.1082×10^{-4}	0.1002	0.1096×10^{-4}	0.0187	0.3202×10^{-5}	0.0104	0.2839×10^{-5}
		-0.0490	0.3391	0.0636	0.7149×10^{-1}	0.2004	0.1150×10^{-1}	0.0085	0.1404×10^{-2}	0.0113	0.3121×10^{-2}
4		0.3693	0.1624×10^{1}	0.0800	0.1515×10^{-1}	0.0400	0.4995×10^{-3}	0.0205	0.2327×10^{-2}	0.0306	0.4941×10^{-1}
	16	-0.0003	0.7301×10^{-3}	0.0000	0.1011×10^{-4}	0.0001	0.1079×10^{-4}	-0.0000	0.3051×10^{-5}	0.0002	0.2726×10^{-5}
		0.1141	0.7301×10^{-3}	0.0210	0.1011×10^{-4}	0.1016	0.1079×10^{-4}	0.0194	0.3051×10^{-5}	0.0102	0.2726×10^{-5}
		-0.0047	0.4140	0.0638	0.8396×10^{-1}	0.2036	0.3485×10^{-1}	0.0090	0.7298×10^{-2}	0.0107	0.1133×10^{-1}

TABLE 3.16

DATA SET 1

MODEL	RUN NO	$(\Sigma^{N+1})^{-1}$	$\lvert\Sigma^{N+1}\rvert$	x_1	x_2	x_3	Prior Prob	Post.Prob
	16	57968-3.4902-3.4902-3.4873	$.49471\times10^{-5}$	.001	29.999	.001	.25000	.47380
	17	61229-10.508-10.508-10.505	$.15556\times10^{-5}$	.000	29.999	3.099	.47380	.48411
1	18	45986-12.766-12.766-12.765	$.17040\times10^{-5}$	.001	29.999	18.579	.48411	.47168
	19	48633-16.118-16.118-16.116	$.12763\times10^{-5}$	.001	29.999	18.578	.47168	.46567
	20	6278.3 8.8405 8.8405 13.622	$.11704\times10^{-4}$	9.222	.001	29.999	.46567	.40866
	16	116.78 2.4443 2.4443 3.8069	$.22799\times10^{-2}$	.001	29.999	.001	.25000	.02257
	17	13.267 .12709 .12709 10.553	$.71435\times10^{-2}$	.0000	29.999	3.099	.02257	.00035
2	18	9.7795 .03998 .03998 14.329	$.71364\times10^{-2}$	.001	29.999	18.579	.00035	.0
	19	.21830 .00091 .00091 16.829	$.27220\times10^{-0}$	.001	29.999	18.578	0.0	.0
	20	6.4308 5.3213 5.3213 8.4670	$.38266\times10^{-1}$	9.222	.001	29.999	.0	.0
	16	57914-3.8365-3.8365 3.8383	$.45041\times10^{-5}$	.001	29.999	.001	.25000	.49660
	17	61147-10.663-10.663 10.661	$.15343\times10^{-5}$	.0000	29.999	3.099	.49660	.51084
	18	45918 -14.402-14.402 14.401	$.15128\times10^{-5}$	.001	29.999	18.579	.51084	.52824
	19	48598 -16.931-16.931 16.930	$.17159\times10^{-5}$	.001	29.999	18.578	.52824	.53433
	20	6200.5 17.178 17.178 23.537	$.68660\times10^{-5}$	9.222	.001	29.999	.53433	.59134
	16	14.921 2.4545 2.4545 3.2220	$.23781\times10^{-1}$	.001	29.999	.001	.25000	.00702
	17	13.920 .17257 .17257 10.324	$.69597\times10^{-2}$	.0000	29.999	3.099	.00702	.00470
	18	11.287 .08125 .08125 12.564	$.70517\times10^{-2}$	.001	29.999	18.579	.00478	.0
	19	.28734 .00116 .00116 15.936	$.21838\times10^{0}$	.001	29.999	18.573	.0	.0
	20	2.1833 1.7294 1.7294 4.7065	.13727	9.222	.001	29.999	.0	.0

DATA SET 2

MODEL	RUN NO	$(\Sigma^{N+1})^{-1}$	$\lvert\Sigma^{N+1}\rvert$	x_1	x_2	x_3	Prior Prob	Post. Prob
	16	21538-3.4214-3.4214 3.4187	$.13583 \times 10^{-4}$	.001	29.999	.001	.25000	.47686
	17	22765-10.350-10.350-10.349	$.42464 \times 10^{-5}$	.001	29.999	17.382	.47686	.49899
1	18	14022-11.889-11.889-11.889	$.60039 \times 10^{-5}$	.001	29.999	29.637	.49899	.47703
	19	14839-13.842-13.842 13.842	$.48732 \times 10^{-5}$	.001	29.999	.001	.47703	.45332
	20	2600.2 7.9360 7.9360 14.054	$.27411 \times 10^{-4}$	25.713	.001	29.999	.45332	.38539
	16	49.715 2.2930 2.2930 3.6646	$.56521 \times 10^{-2}$	.001	29.999	.001	.25000	.01669
	17	14.732 .11790 .11790 10.476	$.64803 \times 10^{-2}$	.001	29.999	17.382	.01669	.00045
2	18	11.104 .02945 .02945 14.179	$.63516 \times 10^{-2}$	.001	29.999	29.637	.00045	.0
	19	.22299 .000416.000416 16.672	$.26898 \times 10^{0}$	.001	29.999	.001	.0	.0
	20	7.2372 5.5206 5.5206 9.7289	$.25042 \times 10^{-1}$	25.713	.001	29.999	.0	.0
	16	20882-3.7275-3.7275 3.7251	$.12858 \times 10^{-4}$	.001	29.999	.001	.25000	.49095
	17	20990-10.579-10.579 10.579	$.45057 \times 10^{-5}$	.001	29.999	17.382	.49095	.50005
3	18	13992-14.260-14.260 14.259	$.50173 \times 10^{-5}$	.001	29.999	29.637	.50005	.57293
	19	14816-16.769-16.769 16.768	$.40296 \times 10^{-5}$	.001	29.999	.001	.52293	.54668
	20	2517.9-14.708-14.708 23.212	$.17174 \times 10^{-4}$	25.713	.001	29.999	.54668	.61461
	16	34.436 2.4980 2.4980 3.3155	$.92650 \times 10^{-2}$	.001	29.999	.001	.25000	.01551
	17	22.387 .17814 .17814 10.222	$.43704 \times 10^{-2}$	.001	29.999	17.382	.01551	.00051
4	18	82.723 .03057 .03057 11.837	$.10212 \times 10^{-2}$	.001	29.999	29.637	.00051	.0
	19	.14295-.00013-.00013 13.820	$.50619 \times 10^{0}$	.001	29.999	.001	.0	.0
	20	4.2141 2.9504 2.9504 6.6977	$.51231 \times 10^{-1}$	25.713	.001	29.999	.0	.0

TABLE 3.18

DATA SET 3

MODEL	RUN NO	$(\Sigma^{N+1})^{-1}$	$\|\Sigma^{N+1}\|$	x_1	x_2	x_3	Prior Prob	Post Prob
1	16	4858.3 -2.9636 -2.9636 -2.9600	$.69581\times10^{-4}$	.001	29.999	.001	.25000	.47383
	17	5175.9 -6.3763 -6.3763 6.3755	$.30341\times10^{-4}$	.001	29.999	29.999	.47383	.46887
	18	5384.3 -10.605 -10.605 10.603	$.17550\times10^{-4}$	.001	29.999	3.470	.46887	.47485
	19	5661.5 -12.378 -12.378 12.377	$.14303\times10^{-4}$	.001	29.999	29.999	.47485	.47805
	20	176.78 12.990 12.990 13.767	$.44151\times10^{-3}$	29.999	7.132	29.999	.47805	.36928
2	16	3.1660 1.1109 1.1109 2.3810	$.15863\times10^{0}$	.001	29.999	.001	.25000	.00995
	17	14.140 .08707 .08707 8.4683	$.83520\times10^{-2}$	.001	29.999	29.999	.00995	.00059
	18	10.650 .01084 .01084 11.366	$.82609\times10^{-2}$	.001	29.999	3.470	.00059	.0
	19	.22060 .00021 .00021 13.338	.33986	.001	29.999	29.999	.0	.0
	20	6.1847 5.9379 5.9379 6.4402	.21868	29.999	7.132	29.999	.0	.0
3	16	4644.1 -3.0652 -3.0652 3.0619	$.70372\times10^{-4}$	.001	29.999	.001	.25000	.47146
	17	4932.0 -8.5755 -8.5755 8.5743	$.23688\times10^{-4}$	.001	29.999	29.999	.47146	.52816
	18	4802.3 11.458 11.458 11.457	$.18218\times10^{-4}$	.001	29.999	3.470	.52316	.52511
	19	5085.7 13.434 13.434 13.433	$.14676\times10^{-4}$	.001	29.999	29.999	.52511	.52195
	20	160.02 17.381 17.381 18.401	$.37844\times10^{-3}$	29.999	7.132	29.999	.52195	.63072
4	16	73.411 1.4940 1.4940 2.9308	$.46965\times10^{-2}$	.001	29.999	.001	.25000	.04475
	17	15.064 .10623 .10623 6.2757	$.10579\times10^{-1}$	.001	29.999	29.999	.04475	.00238
	18	.11063 .00052 .00052 10.491	$.86161\times10^{0}$	.001	29.999	3.470	.00238	.0
	19	.34299 .00058 .00058 12.275	.23751	.001	29.999	29.999	.0	.0
	20	.11599 .11185 .11185 .60843	$.1722\times10^{-2}$	29.999	7.132	29.999	.0	.0

TABLE 3.19

DATA SET 4

MODEL	RUN NO	$(\Sigma^{N+1})^{-1}$	$\lvert\Sigma^{N+1}\rvert$	x_1	x_2	x_3	Prior Prob	Post Pr
	16	2299.1 21.198 21.198 20.771	$.21139 \times 10^{-4}$	29.999	.001	29.999	.25000	.37045
1	17	88668-.06559-.06559 .06539	$.17247 \times 10^{-3}$	.001	29.999	10.577	.37045	.48678
	18	83251-.44635-.44635 .44657	$.26898 \times 10^{-4}$	.001	29.999	13.073	.48678	.58040
	19	47459-.60737-.60737 .60470	$.18909 \times 10^{-4}$	3.904	29.999	13.750	.58040	.66359
	20							
	16	96.493 36.891 36.891 37.332	$.44616 \times 10^{-3}$	29.999	.001	29.999	.25000	.08502
2	17	456.14-.00189-.00189 .03693	$.59355 \times 10^{-1}$	.001	29.999	10.577	.08502	.00619
	18	14.537 .00007 .00007 .23955	.28720	.001	29.999	13.073	.00619	.0
	19	10.441 .00001 .00001 .32854	.29151	3.904	29.999	13.750	.0	.0
	20							
	16	.2165.1 35.514 35.514 37.571	$.12487 \times 10^{-4}$	29.999	.001	29.999	.25000	.52879
3	17	82542-.03721-.03721-.03694	$.32800 \times 10^{-3}$	.001	29.999	10.577	.52879	.50534
	18	74929-.24091-.24091 .23955	$.55713 \times 10^{-4}$	.001	29.999	13.073	.50534	.41952
	19	78846-.33041-.33041 .32854	$.38604 \times 10^{-4}$	3.904	29.999	13.750	.41952	.33641
	20							
	16	24.022 19.835 19.835 19.333	$.14088 \times 10^{-1}$	29.999	.001	29.999	.25000	.01575
4	17	584.62-.00579-.00579 .06539	$.26159 \times 10^{-1}$	.001	79.999	10.577	.01575	.00168
	18	1.2388 .00144-.00144 .44346	1.8203	.001	29.999	13.073	.00168	.0
	19	131.84-.00260-.00260 .60464	$.12544 \times 10^{-1}$	3.904	29.999	13.750	.0	.0
	20							

TABLE 3.20

DATA SET 5

MODEL	RUN NO	$(\Sigma^{N+1})^{-1}$				$\lvert \Sigma^{N+1} \rvert$	x_1	x_2	x_3	Prior Prob	Post Pro
1	16	1323.6	-0.00353	-0.00353	.00354	$.21370\times10^{0}$	.001	29.999	29.999	.25000	.17370
	17	1371.3	-.07329	-.07329	.07384	$.98759\times10^{-2}$	.00969	3.1130	29.308	.17370	.05068
2	16	.76478	.02753	.02753	.07761	$.17064\times10^{2}$	.001	29.999	29.999	.25000	.01712
	17	305.16	-.29344	-.29344	1.2399	$.26436\times10^{-2}$	.00969	3.1130	29.308	.01712	.00914
3	16	1265.9	-.07959	-0.07959	.07955	$.99315\times10^{-2}$	.001	29.999	29.999	.25000	.80641
	17	1302.8	-1.2408	-1.2408	1.2408	$.61919\times10^{-3}$	.00969	3.1130	29.308	.80641	.94016
4	16	.39921	-.00409	-.00409	.00345	$.73415\times10^{3}$	.001	29.999	29.999	.25000	.00277
	17	.60957	-.00073	-.00073	.04660	$.35199\times10^{2}$	.00969	3.1130	29.308	.00277	.00001

TABLE 3.21

DATA SET 6

DEL	RUN NO	$(\Sigma^{N+1})^{-1}$				$\lvert\Sigma^{N+1}\rvert$	x_1	x_2	x_3	Prior Prob	Post.Prob
1	16	20181	.22244	.22244	.04390	$.11288\times10^{-2}$	29.999	.001	29.999	.25000	.92586
	17	98655	-6.1853	-6.1853	6.2064	$.16333\times10^{-5}$	.001	29.999	29.999	.92586	.99081
2	16	101.57	9.4871	9.4871	12.602	$.84040\times10^{-3}$	29.999	.001	29.999	.25000	0.00000
	17	15.619	-.55608	-.55608	.59843	$.11065\times10^{0}$	.001	29.999	29.999	0.00000	0.00000
3	16	328.23	7.0964	7.0964	12.797	$.24096\times10^{-3}$	29.999	.001	29.999	.25000	0.00000
	17	23.001	-.59949	-.59949	.59947	$.74466\times10^{-1}$	.001	29.999	29.999	.00000	0.00000
4	16	120.89	.14048	.14048	.04386	$.18930\times10^{0}$	29.999	.001	29.999	.25000	0.07414
	17	46.555	.46356	.46356	5.8736	$.36599\times10^{-2}$	.001	29.999	29.999	0.07414	0.00199

TABLE 3.22

DATA SET 7

MODEL	RUN NO	$(\Sigma^{N+1})^{-1}$	$\lvert\Sigma^{N+1}\rvert$	x_1	x_2	x_3	Prior Prob.	Post. Prob
1	16	36484 1.5916 1.5916 .08081	$.33948x10^{-3}$	29.999	.001	30.00	.25000	.94570
	17	92814-15.280-15.280 15.283	$.70512x10^{-6}$	.001	29.999	.001	.94570	.93291
2	16	36.339 18.232 18.232 30.928	$.12634x10^{-2}$	29.999	.001	30.000	.25000	0.00000
	17	.98134-.18592-.18592 .20464	$.60149x10^{1}$	.001	29.999	.001	.00000	0.00000
3	16	506.64 7.2225 7.2225 43.192	$.45808x10^{-4}$	29.999	.001	30.000	.25000	0.00000
	17	1.6887-.20529-.20529 .20528	$.32838x10^{1}$	.001	29.999	.001	.00000	0.00000
4	16	122.14 .95826 .95826 .07742	$.11713x10^{0}$	29.999	.001	30.000	.25000	0.05430
	17	1053.4-.04156-.04156 15.277	$.62140x10^{-4}$	.001	29.999	.001	.00000	.00709

TABLE 3.23

DATA SET 8

MODEL	RUN NO	$(\Sigma^{N+1})^{-1}$				$\lvert\Sigma^{N+1}\rvert$	x_1	x_2	x_3	Prior Prob.	Post Prob.
		204030	16.601	16.601	3.6919	$.13281\times10^{-5}$	17.164	11.897	23.085	.25000	.94279
		367230	-32.235	-32.235	32.208	$.8455\times10^{-7}$	0.001	29.999	29.999	.94279	.99959
2	16	318.21	20.675	20.675	78.958	$.40490\times10^{-4}$	17.164	11.897	23.005	.25000	.02966
	17	13.509	3.612	3.612	14.398	$.5510\times10^{-2}$	0.001	29.999	29.999	.02966	.00019
3	16	47454	-43.722	-43.722	83.173	$.25349\times10^{-6}$	17.164	11.897	23.085	.25000	.00676
	17	535.54	-19.485	-19.485	19.484	$.9945\times10^{-4}$	0.001	29.999	29.999	.00676	.00019
4	16	101.68	4.6672	4.6672	1.9438	$.56863\times10^{-2}$	17.164	11.897	23.085	.25000	.02079
	17	8.182	3.668	3.668	16.273	$.8353\times10^{-2}$	0.001	29.999	29.999	.02079	.00011

TABLE 3.24
DATA SET 9

MODEL	RUN NO	$(\Sigma^{N+1})^{-1}$				$\lvert\Sigma^{N+1}\rvert$	x_1	x_2	x_3	Prior Prob.	Post Prob.
	16	312390	-82.666	-82.666	82.639	$.38747 \times 10^{-7}$	.001	29.999	27.757	.25000	.70482
1	17	94369	-3.182	-3.182	.759	$.13950 \times 10^{-4}$	29.999	0.001	29.999	.70482	.99874
	16	29.745	9.1233	9.1233	20.895	$.18577 \times 10^{-2}$	.001	29.999	27.757	.25000	.00521
2	17	274.53	68.446	68.446	99.045	$.4443 \times 10^{-4}$	29.999	0.001	29.999	.00521	.00016
	16	70414	-26.106	-26.106	26.101	$.54432 \times 10^{-6}$	.001	29.999	27.757	.25000	.28663
3	17	8822.5	45.601	45.601	108.88	$.10433 \times 10^{-5}$	29.999	0.001	29.999	.28663	.00088
	16	10.588	6.8486	6.8486	22.400	$.52558 \times 10^{-2}$	.001	29.999	27.757	.25000	.00335
4	17	140.21	-1.149	-1.149	.754	$.9567 \times 10^{-2}$	29.999	.001	29.999	.00335	.00020

to be nearly 10^3 times greater than the remaining elements and also the estimated value for the (2,2) element was not in good agreement with the value used for data generation.

3.3 Sequential Design of Experiments.

After obtaining the estimated values of $\hat{\theta}$ and $\hat{\Sigma}r$ the scalar discriminant function Kv was maximized using Box's[25] algorithm.

The elements of $(\Sigma r^{N+1})^{-1}$ formed corresponding to the highest value of Kv within the operability region are reported in Tables 3.16 through 3.24. Also the values of the independent variables x_1, x_2, x_3 namely concentrations of component A,B and C are also presented in those tables. These values of the independent variables were used for conducting the $(N+1)^{th}$ run i.e. the next experiment and the values of the responses were simulated using the same strategy as that of the original data generation. These concentration data with the responses for different sets are shown in Tables 3.1 through 3.9.

3.4 Posterior Probability.

After conducting the $(N+1)^{th}$ run the posterior probabilities of all the models were calculated using equation 2.5.1 and equation 2.3.13.

If the posterior probability was below .9 for the models the whole sequence from 3.2 to 3.4 was repeated and the probabilities are reported in Tables 3.16 through 3.24.

An overall review of the Tables 3.16 through 3.24 indicates that

(i) For data sets 1,2,3 the posterior probabilities for the correct models (i.e. Model 3) showed an increase from .25 to 0.48 nearly after

sequential design. In the subsequent runs the probabilities increased gradually but with some oscillations to values round about 0.6.

(ii) For data set 4, the posterior probability for the correct model (model 3) increased from .25 to .52 after the 16th run. However during the 19th run the probability reduced to 0.33(nearly) and further calculation was stopped. Explanation is given in the next paragraph.

(iii) For data sets 5,6,7,8,9 after the 16th run i.e. for the 1st sequential run the posterior probabilities for the correct model showed a rapid increase from the initial model probability. That is from the initially assigned model probability of .25 it went as high as .8 to .9. After 17th run the probabilities of the correct model approached 1 and further calculation was not necessary.

The expression that is used for finding the posterior probabilities of the models is given by equation 2.5.1 where the likelihood for the models after the $(N+1)^{th}$ run is calculated using equation 2.3.13. The most dominant factor in equation 2.3.13 was the exponential factor and the exponent on this factor contains expression involving $(\Sigma_r^{N+1})^{-1}$ and $(Y^{N+1} - \hat{Y}_r^{N+1})$. From the tables 3.16 through 3.24 it can be seen that the (Σ_r^{N+1}) matrices are ill conditioned for most of the cases, as indicated by the small values of the determinant for Σ_r^{N+1}. Under this circumstances, if the difference $(Y^{N+1} - \hat{Y}_r^{N+1})$ is not very small, the likelihood for the models approaches very small values. However, in the present study the error added for the correct model was small in magnitude and didnot pose any serious problems for discrimination purpose.

Looking into the posterior probability values for data set 1,2 and 3, it can be seen that model 1 and model 3 (model 3 is the correct model) are competing whereas the remaining model probabilities approache very small values. The main reason is that during parameter estimation the parameter θ_{13} for the 1st model approached zero and virtually there was no difference between model 1 and model 3 except for the estimates of variance covariance matrix for obvious reasons and consequently the difference between $(Y^{N+1} - \hat{Y}_r^{N+1})$ was much smaller for model 1 and 3 compared to model 2 and 4. These factors account for the probability values as obtained which is expected. Therefore Box and Hill's criterion was adequate enough to discriminate between rival models for data set 1,2 and 3.

For data set 4 from Table 3.19 it is seen that the (1,1) element of $(\sum_Y^{N+1})^{-1}$ for the 19th run was much smaller for model 1 compared to model 3. Also the values for $(Y^{N+1} - \hat{Y}_r^{N+1})$ for both models are comparable with each other as explained above, and hence model 1 (the incorrect model) prevailed over model 3 (the correct model).

For data sets 6,7,8 and 9 as already pointed out, the values of the model probabities showed a large change after the 1st sequential run. The reason behind this is that the difference $(Y^{N+1} - \hat{Y}_r^{N+1})$ was significant for all the models except model 1 (the correct model) and hence the likelihood values for model 1 must. have taken larger magnitudes compared to other models accounting for the very high posterior probabilities as shown in the Tables.

For data set 5, the correct model is 3 and the same analysis as given for data sets 6,7,8,9 holds good in this case.

Here it should also be pointed out that in the expression for K_v, equation 2.3.18 , though the prior probabilities of the models are used as weights but as in the present case where the $(\Sigma_r^{N+1})^{-1}$ has large elements, the weights attached can be offset by the remaining factors containing $(\Sigma_r^{N+1})^{-1}$ terms. This may lead to inefficient design and may cause fluctuations in the posterior probability values.

The probability density function of $\hat{Y}_r^{N+1}$ given Σ_r is

$$p\,(Y_r^{N+1} \mid \Sigma_r) = \frac{\left|\Sigma_r^{N+1}\right|^{-\frac{1}{2}}}{(2\pi)^u} \exp\left[-\tfrac{1}{2}\,(Y^{N+1} - \hat{Y}_r^{N+1})^T \right.$$

$$\left. (\Sigma_r^{N+1})^{-1}\,(Y^{N+1} - \hat{Y}_r^{N+1})\right] \quad \ldots(4.1)$$

where $\Sigma_r^{N+1} = \Sigma_r + W_r^{N+1}$ .... (4.2)

If the marginal probability density function of Y_r^{N+1} is considered then

$$p\,(Y_r^{N+1}) = \int_0^\infty \cdots \int_0^\infty p\,(Y_r^{N+1} \mid \Sigma_r)\; p\,(\Sigma)\, d\Sigma \quad \ldots\ldots(4.3)$$

In order to find out $p(\Sigma)$ one/use the method suggested by Hill and Hunter (24) for single response case, using non informative prior distribution for σ.

Once $p\,(Y_r^{N+1})$ is obtained from equation 4.3 a criterion similar to Box and Hill[15,16] may be arrived for multiresponse case with unknown variance covariance matrix.

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```
$IBFTC MAIN     NOPRNT
C       MAIN PROGRAM FOR LINEAR REGRESSION IN MULTIRESPONSE SITUATION
        DIMENSION E10(25),SI(4,4),ENSI(2,2)
        COMMON P,Y(50),X(50,4 )/AREA/N,C(3,25),ETA(2,25)/AREA1/S(50,50)
       1 ,B(4)
        COMMON/AREA2/SIG1(2,2),SIG2(2,2),SIG3(2,2),SIG4(2,2)
        INTEGER R,P,U,UU,UU1
C       U               EXPT RUN NO
C       C(1,U)          CONC OF A
C       C(2,U)          CONC OF B
C       C(3,U)          CONC OF C
C       NO OF RESPONSES
C       ETA(1,U)        RATE OF B
C       ETA(2,U)        RATE OF C
C       N               TOTAL NO EXPERIMENTAL RUNS
C       P               NO OF PARAMETERS IN MODELS
C
        R=2
        PRINT 003
003     FORMAT(1H1)
        K=0
        READ 001,IK
C
  10    READ 001,N
001     FORMAT(I2)
        IF(N.EQ.0)  PRINT003
        K=K+1
        N2=2*N
        CONST=1./FLOAT(N)
         READ2  ,((C(J,U),J=1,3),(ETA(I,U),I=1,R),U=1,N)
        PRINT2 ,((C(J,U),J=1,3),(ETA(I,U),I=1,R),U=1,N)
   2    FORMAT(5F7.3)
        PRINT 004,K
004     FORMAT(1H0////49X,31HLINEAR REGRESSION WITH DATA SET,I3/49X,34(1H*
       1))
C
        DO 80 M=1,4
        DO 20  I=1,N2
        DO 20 J=1,N2
        S(I,J)=0.
        IF(I.EQ.J)  S(I,J)=1.
20      CONTINUE
        CALL FORM(M,1)
        CALL LINREG(N)
C
        SI(1,1)=0.
        DO 40 U=1,N
        Y1=0.
        DO 30 J=1,P
        Y1=Y1+X(U,J)*B(J)
 30     CONTINUE
        E10(U)=Y(U)-Y1
        SI(1,1)=SI(1,1)+E10(U)*E10(U)
   40   CONTINUE
        SI(1,1)=CONST*SI(1,1)
        CALL FORM(M,2)
        CALL LINREG(N)
        SI(1,2)=0.
        SI(2,2)=0.
        DO 60 U=1,N
        Y1=0.
        DO 50 J=1,P
        Y1=Y1+X(U,J)*B(J)
 50     CONTINUE
        E20=Y(U)-Y1
        SI(1,2)=SI(1,2)+E10(U)*E20
```

```
      SI(2,2)=SI(2,2)+E20*E20
60    CONTINUE
      SI(1,2)=CONST*SI(1,2)
      SI(2,1)=SI(1,2)
      SI(2,2)=CONST*SI(2,2)
      PRINT 555,((SI(I,J),J=1,2),I=1,2)
      PUNCH 555,((SI(I,J),J=1,2),I=1,2)
  555 FORMAT(4E12.5)
  444 FORMAT(25(2H**))
C     SI ESTIMATED VAR-COVAR MATRIX OF OBS VECTOR IN EACH RUN
      CALL INVS(SI,2)
      GO TO (61,62,63,64),M
   61 DO 66 I1=1,2
      DO 66 J1=1,2
   66 SIG3(I1,J1)=SI(I1,J1)
      GO TO 65
   62 DO 67 I2=1,2
      DO 67 J2=1,2
   67 SIG1(I2,J2)=SI(I2,J2)
      GO TO 65
   63 DO 68 I3=1,2
      DO 68 J3=1,2
   68 SIG2(I3,J3)=SI(I3,J3)
      GO TO 65
   64 DO 69 I4=1,2
      DO 69 J4=1,2
   69 SIG4(I4,J4)=SI(I4,J4)
   65 PRINT444
      DO 70 U=1,N
      UU=2*U-1
      UU1=UU+1
      S(UU,UU)=SI(1,1)
      S(UU1,UU)=SI(2,1)
      S(UU,UU1)=SI(1,2)
      S(UU1,UU1)=SI(2,2)
70    CONTINUE
C
      CALL FORM(M,0)
      CALL LINREG(N2)
      PRINT 007,  (B(I),I=1,P)
007    FORMAT (//,50X,3X,4F12.4)
      PUNCH 555,(B(I),I=1,P)
80    CONTINUE
      IF(K/3*3.EQ.K) PRINT 003
      CALL MATRIX(N,C)
      IF(K.EQ.IK) GO TO 900
      GO TO 10
900   STOP
      END
$IBFTC FORM    NOPRNT
      SUBROUTINE FORM(M,I)
C  THIS SUB FORMS THE OBS VECTOR YANDDESIGN MATRIX X FOR MODEL M
      COMMON P,Y(50),X(50,4 )/AREA/N,C(3,25),ETA(2,25)
      INTEGER P,U,UN
      P=M+(6-M)/4
      IF (I.EQ.0) GO TO 100
      GO TO (10,70),I
10      DO 20 U=1,N
      Y(U)=ETA(1,U)
      X(U,1)=C(1,U)
      X(U,2)=-C(2,U)
20    CONTINUE
      IF (M.EQ.1) RETURN
      IF (M.GT.2) GO TO 40
      DO 30 U=1,N
      X(U,3)=C(3,U)
30    CONTINUE
```

```
      RETURN
40    DO 50 U=1,N
      X(U,3)=-C(2,U)
50    CONTINUE
      IF (M.EQ.3) RETURN
      DO 60 U=1,N
      X(U,4)=C(3,U)
60    CONTINUE
      RETURN
70    P=P-(M+1)/2
      DO 80 U=1,N
      X(U,1)=C(2,U)
80    CONTINUE
      IF (M/2*2.NE.M) RETURN
      DO 90 U=1,N
      X(U,2)=-C(3,U)
90    CONTINUE
      RETURN
100   DO 110U=1,N
      UN=U+N
      Y(U)=ETA(1,U)
      Y(UN)=ETA(2,U)
      X(U,1)=C(1,U)
      X(UN,1)=0.
      X(U,2)=-C(2,U)
110   CONTINUE
       IF (M.GT.2)  GO TO 140
      DO 120 U=1,N
      UN=U+N
      X(UN,2)=C(2,U)
120   CONTINUE
      IF (M.EQ.1) RETURN
      DO 130 U=1,N
      UN=U+N
      X(U,3)=C(3,U)
      X(UN,3)=-C(3,U)
130   CONTINUE
      RETURN
140   DO 150 U=1,N
      UN=U+N
      X(UN,2)=0.
      X(U,3)=-C(2,U)
      X(UN,3)=C(2,U)
150   CONTINUE
      IF (M.EQ.3) RETURN
      DO 160 U=1,N
      UN=U+N
      X(U,4)=C(3,U)
      X(UN,4)=-C(3,U)
160   CONTINUE
      RETURN
      END
$IBFTC LINREG  NOPRNT
      SUBROUTINE LINREG(N)
C     FOR LINEAR REGRESSION WHICH RETURNS THE VECTOR OF P EST PARA
C     METERS
      COMMON P,Y(50),X(50,4 )/AREA1/S(50,50),B(4)
      INTEGER P
      DIMENSION XT(4,50),XTS(4,50),XTSX(4,4),XTSY(4)
      DO 10 I=1,P
      DO 10 J=1,N
      XT(I,J)=X(J,I)
10    CONTINUE
C     S    INVERSE OF VAR COVAR MATRIX OF OBS VECTOR Y
      DO 20 I=1,P
      DO 20 J=1,N
      XTS(I,J)=0.
```

```
      DO 20 K=1,N
      XTS(I,J)=XTS(I,J)+XT(I,K)*S(K,J)
20    CONTINUE
      DO 30 I=1,P
      DO 30 J=1,P
      XTSX(I,J)=0.
      DO 30 K=1,N
      XTSX(I,J)=XTSX(I,J)+XTS(I,K)*X(K,J)
30    CONTINUE
      CALL INVS(XTSX,P)
      DO 40 I=1,P
      XTSY(I)=0.
      DO 40 J=1,N
      XTSY(I)=XTSY(I)+XTS(I,J)*Y(J)
40    CONTINUE
      DO 50 I=1,P
      B(I)=0.
      DO 50 J=1,P
      B(I)=B(I)+XTSX(I,J)*XTSY(J)
50    CONTINUE
      RETURN
      END
$IBFTC INVS    NOPRNT
      SUBROUTINE INVS(A,M)
C     SUB FOR INVS OF MAT A
      DIMENSION A(4,4)
      DO 60 K=1,M
      A(K,K)=-1./A(K,K)
      DO 20 I=1,M
      IF(I-K)10,20,10
10    A(I,K)=-A(I,K)*A(K,K)
20    CONTINUE
      DO 40 I=1,M
      DO 40 J=1,M
      IF((I-K)*(J-K)) 30,40,30
30    A(I,J)=A(I,J)-A(I,K)*A(K,J)
40    CONTINUE
      DO 60 J=1,M
      IF(J-K)50,60,50
50    A(K,J)=-A(K,J)*A(K,K)
60    CONTINUE
      DO 70 I=1,M
      DO 70 J=1,M
      A(I,J)=-A(I,J)
70    CONTINUE
      RETURN
      END
$IBFTC  MATRIX NOPRNT
        SUBROUTINE MATRIX(N,X)
      DIMENSION X(3,N),X11(25,3),X12(25,3),XT11(3,25),XT12(3,25),
     1 AMX11(3,3) , AMX12(3,3) , AMX21(3,3) , AMX22(3,3) , MA1(3,3)
     2 , X21(25,3) , X22(25,3),XT21(3,25),XT22(3,25),BMX11(3,3)
     3 , BMX12(3,3) , BMX21(3,3),BMX22(3,3),MA2(3,3),X31(25,2),X32(25,2)
     4 , XT31(2,25),XT32(2,25),CMX11(2,2),CMX12(2,2),CMX21(2,2),
     5 CMX22(2,2),MA3(2,2),X41(25,4),X42(25,4),XT41(4,25),XT42(4,25)
      DIMENSION DMX11(4,4),DMX12(4,4),DMX21(4,4),DMX22(4,4),MA4(4,4),AL1
     1 (3,3),BL1(3,3),CL1(3,3),DL1(3,3),AL2(3,3),BL2(3,3),CL2(3,3),DL2(3
     2 ,3),AL3(2,2),BL3(2,2),CL3(2,2),DL3(2,2),AL4(4,4),BL4(4,4),CL4(4,4
     3 ),DL4(4,4),B1(4,1)
      COMMON/AREA2/SIG1(2,2),SIG2(2,2),SIG3(2,2),SIG4(2,2)
      REAL   MA1,MA2,MA3,MA4
      M1=3
      M2=3
      M3=2
      M4=4
C     MAIN LINE PROGRAM
      WRITE(6,97)((X(I,J),J=1,N),I=1,3)
```

```
 97   FORMAT(15F5.2)
      CALL MFORM(1,X,N,X11,X12,M1)
      CALL TRPOSE(X11,N,M1,XT11)
      CALL TRPOSE(X12,N,M1,XT12)
      CALL MAMUL(XT11,X11,N,M1,AMX11)
      CALL MAMUL(XT11,X12,N,M1,AMX12)
      CALL MAMUL(XT12,X11,N,M1,AMX21)
      CALL MAMUL(XT12,X12,N,M1,AMX22)
      CALL MAT(AMX11,AMX12,AMX21,AMX22,SIG1,M1,MA1,AL1,BL1,CL1,DL1)
      CALL MFORM(2,X,N,X21,X22,M2)
      CALL TRPOSE(X21,N,M2,XT21)
      CALL TRPOSE(X22,N,M2,XT22)
      CALL MAMUL(XT21,X21,N,M2,BMX11)
      CALL MAMUL(XT21,X22,N,M2,BMX12)
      CALL MAMUL(XT22,X21,N,M2,BMX21)
      CALL MAMUL(XT22,X22,N,M2,BMX22)
      CALL MAT(BMX11,BMX12,BMX21,BMX22,SIG2,M2,MA2,AL2,BL2,CL2,DL2)
      CALL MFORM(3,X,N,X31,X32,M3)
      CALL TRPOSE(X31,N,M3,XT31)
      CALL TRPOSE(X32,N,M3,XT32)
      CALL MAMUL(XT31,X31,N,M3,CMX11)
      CALL MAMUL(XT31,X32,N,M3,CMX12)
      CALL MAMUL(XT32,X31,N,M3,CMX21)
      CALL MAMUL(XT32,X32,N,M3,CMX22)
      CALL MAT(CMX11,CMX12,CMX21,CMX22,SIG3,M3,MA3,AL3,BL3,CL3,DL3)
      CALL MFORM(4,X,N,X41,X42,M4)
      CALL TRPOSE(X41,N,M4,XT41)
      CALL TRPOSE(X42,N,M4,XT42)
      CALL MAMUL(XT41,X41,N,M4,DMX11)
      CALL MAMUL(XT41,X42,N,M4,DMX12)
      CALL MAMUL(XT42,X41,N,M4,DMX21)
      CALL MAMUL(XT42,X42,N,M4,DMX22)
      CALL MAT(DMX11,DMX12,DMX21,DMX22,SIG4,M4,MA4,AL4,BL4,CL4,DL4)
      CALL MATINV(MA1,3,B1,0,DETER )
      CALL MATINV(MA2,3,B1,0,DETER)
      CALL MATINV(MA3,2,B1,0,DETER)
      CALL MATINV(MA4,4,B1,0,DETER)
      WRITE(6,333) ((MA1(I,J),J=1,3),I=1,3)
      WRITE(6,333) ((MA2(I,J),J=1,3),I=1,3)
      WRITE(6,333) ((MA3(I,J),J=1,2),I=1,2)
      WRITE(6,333) ((MA4(I,J),J=1,4),I=1,4)
      PUNCH 333,((MA1(I,J),J=1,3),I=1,3)
      PUNCH 333,((MA2(I,J),J=1,3),I=1,3)
      PUNCH 333,((MA3(I,J),J=1,2),I=1,2)
      PUNCH 333,((MA4(I,J),J=1,4),I=1,4)
 333  FORMAT(6E12.5)
      WRITE(6,999)
 999  FORMAT(50(2H**))
      RETURN
      END
C
$IBFTC MFORM   NOPRNT
      SUBROUTINE MFORM(IMODEL,X,N,Z1,Z2,M)
      DIMENSION X(3,N),Z1(N,M),Z2(N,M)
      IF(IMODEL.NE.1) GO TO 100
      DO 10 J=1,N
      Z1(J,1)=-X(1,J)
      Z1(J,2)=0.
      Z1(J,3)=0.
      Z2(J,1)=X(1,J)
      Z2(J,2)=-X(2,J)
 10   Z2(J,3)=X(3,J)
 100  IF( IMODEL.NE.2) GO TO 101
      DO 11 J=1,N
      Z1(J,1)=-X(1,J)
      Z1(J,2)=X(2,J)
      Z1(J,3)=0.
```

```
      Z2(J,1)=X(1,J)
      Z2(J,2)=-X(2,J)
 11   Z2(J,3)=-X(2,J)
 101  IF (IMODEL.NE.3) GO TO 102
      DO 12 J=1,N
      Z1(J,1)=-X(1,J)
      Z1(J,2)=0.
      Z2(J,1)=X(1,J)
      Z2(J,2)=-X(2,J)
 12   CONTINUE
 102  IF (IMODEL.NE.4) GO TO 103
      DO 13 J=1,N
      Z1(J,1)=-X(1,J)
      Z1(J,2)=X(2,J)
      Z1(J,3)=0.
      Z1(J,4)=0.
      Z2(J,1)=X(1,J)
      Z2(J,2)=-X(2,J)
      Z2(J,3)=-X(2,J)
 13   Z2(J,4)=X(3,J)
 103  RETURN
      END
C
$IBFTC TRPOSE  NOPRNT
      SUBROUTINE TRPOSE(INMAT,IN,IM,OUTMAT)
      DIMENSION INMAT(IN,IM),OUTMAT(IM,IN)
       REAL INMAT
      DO 10 I=1,IN
      DO 10 J=1,IM
 10   OUTMAT(J,I)=INMAT(I,J)
      RETURN
      END
C
$IBFTC MAMUL   NOPRNT
      SUBROUTINE MAMUL(A,B,IN,IM,C)
      DIMENSION A(IM,IN),B(IN,IM),C(IM,IM)
      DO 10 I=1,IM
      DO 10 J=1,IM
      C(I,J)=0.
      DO 10 K=1,IN
 10   C(I,J)=C(I,J)+A(I,K)*B(K,J)
      RETURN
      END
C
$IBFTC MAT     NOPRNT
      SUBROUTINE MAT(A,B,C,D,SIGMA,M,MA,AL,BL,CL,DL)
      DIMENSION A(M,M),B(M,M),C(M,M),D(M,M),AL(M,M),BL(M,M),CL(M,M),DL(M
     1,M),SIGMA(2,2),MA(M,M)
      REAL MA
      DO 10 I=1,M
      DO 10 J=1,M
      AL(I,J)=SIGMA(1,1)*A(I,J)
      BL(I,J)=SIGMA(1,2)*B(I,J)
      CL(I,J)=SIGMA(2,1)*C(I,J)
      DL(I,J)=SIGMA(2,2)*D(I,J)
 10    MA(I,J)=AL(I,J)+BL(I,J)+CL(I,J)+DL(I,J)
      RETURN
      END
$IBFTC MATINV  NOPRNT
      SUBROUTINE MATINV(A,N,B,M,DETERM)
       DIMENSION A(N,N),B(N,M),IPIVOT(40),INDEX(40,2)
      EQUIVALENCE (IROW,JROW), (ICOLUM,JCOLUM), (AMAX,T,SWAP)
 10   DETERM=1.0
 15   DO 20 J=1,N
 20   IPIVOT(J)=0
C     SEARCH FOR PIVOT ELEMENT
 30   DO 550 I=1,N
```

```
 40  AMAX=0.0
 45  DO 105 J=1,N
 50  IF(IPIVOT(J)-1) 60,105 ,60
 60  DO 100 K=1,N
 70  IF(IPIVOT(K)-1) 80, 100, 740
 80  IF(AMAX-ABS(A(J,K))) 85,100, 100
 85  IROW=J
 90  ICOLUM=K
 95  AMAX=ABS(A(J,K))
100  CONTINUE
105  CONTINUE
110  IPIVOT(ICOLUM )=IPIVOT(ICOLUM )+1
C    INTERCHANGE ROWS TO PUT PIVOT ELEMENT ON DIAGONAL
130  IF(IROW-ICOLUM ) 140, 260,140
140  DETERM=-DETERM
150  DO 200 L=1,N
160  SWAP=A(IROW,L)
170  A(IROW,L)=A(ICOLUM ,L)
200  A(ICOLUM ,L)=SWAP
205  IF(M) 260, 260,210
210  DO 250 L=1,M
220  SWAP=B(IROW,L)
230  B(IROW,L)=B(ICOLUM ,L)
250  B(ICOLUM ,L)=SWAP
260  INDEX(I,1)=IROW
270  INDEX(I,2)=ICOLUM
C    DIVIDE PIVOT ROW BY PIVOT ELEMENT
310  PIVOT =A(ICOLUM ,ICOLUM )
320  DETRM=DETERM*PIVOT
330  A(ICOLUM,ICOLUM)=1.0
340  DO 350 L=1,N
350  A(ICOLUM,L)=A(ICOLUM,L)/PIVOT
355  IF(M) 380, 380, 360
360  DO 370 L=1,M
370  B(ICOLUM,L)=B(ICOLUM,L)/ PIVOT
C    REDUCE NON PIVOT ROWS
380  DO 550 L1=1,N
390  IF(L1-ICOLUM) 400, 550,400
400  T=A(L1,ICOLUM)
420  A(L1,ICOLUM)=0.0
430  DO 450 L=1,N
450  A(L1,L)=A(L1,L)-A(ICOLUM,L)*T
455  IF(M) 550, 550, 460
460  DO 500 L=1,M
500  B(L1,L)=B(L1,L)-B(ICOLUM,L)*T
550  CONTINUE
C    INTERCHANGE COLUMS
600  DO 710 I=1,N
610  L=N+1-I
620  IF(INDEX(L,1)-INDEX(L,2)) 630, 710, 630
630  JROW=INDEX(L,2)
650  DO 705 K=1,N
660  SWAP=A(K,JROW)
670  A(K,JROW)=A(K,JCOLUM)
700  A(K,JCOLUM)=SWAP
705  CONTINUE
710  CONTINUE
     DO 11 K=1,N
     IF(IPIVOT(K).NE.1) GO TO 12
 11    CONTINUE
     RETURN
 12  PRINT 991
991  FORMAT(/30X*MATRIX IS SINGULAR*/)
740  RETURN
     END
$ENTRY
```

```
$WATFOR MEF287
$IBJOB
$IBFTC MAIN
C       MAIN LINE PROGRAM FOR COMPLEX ALGORITHM OF BOX
        DIMENSION X(6,4),R(6,3),F(6),G(4),H(4),XC(3)
        COMMON THETA(4,4),P(4),OSI(4,4),DET1,DET2,DET3,DET4,
       1              A(3,3),B(2,2),C(3,3),D(2,2),E(2,2),AF(2,2),AG(4,4),
       2 AH(2,2)
        INTEGER GAMMA
        N1=3
        N2=3
        N3=2
        N4=4
        N5=4
        N6=4
        N7=2
        NMODEL=4
C
        NI=5
        NO=6
C
        READ(5 ,1)N,M,K,ITMAX,IC,IPRINT
    1    FORMAT(8I5)
        READ(5 ,2) ALPHA,BETA,GAMMA
    2    FORMAT(2E10.4,I5)
        DELTA=0.001
    4    FORMAT(8E10.4)
        DO 101 II=2,K
        READ(5 ,3) (R(II,JJ),JJ=1,N)
   3    FORMAT(3F8.4)
  101   CONTINUE
        DO 399 IDIF=1,9
        WRITE(6,104) IDIF
  104   FORMAT(1X,*DATA SET *,I2)
         READ(5 ,100)((A(I,J),J=1,N1),I=1,N1)
         READ(5 ,100)((B(I,J),J=1,N7),I=1,N7)
         READ(5 ,100)((C(I,J),J=1,N2),I=1,N2)
        READ(5  ,100)((D(I,J),J=1,N7),I=1,N7)
         READ(5 ,100)((E(I,J),J=1,N3),I=1,N3)
         READ(5 ,100)((AF(I,J),J=1,N7),I=1,N7)
        READ(5,100) ((AG(I,J),J=1,N4),I=1,N4)
        READ(5,100)((AH(I,J),J=1,N7),I=1,N7)
        READ(5 ,100)(P(I),I=1,NMODEL)
         READ(5,102) ((THETA(I,J),J=1,4),I=1,4)
  100   FORMAT(6E12.5)
  102    FORMAT(4E12.5)
C
        DO 299 ITER =1,2
        READ(5 ,4)(X(1,J),J=1,N)
        WRITE(6,103) (X(1,J),J=1,N)
  103   FORMAT(2X,*INITIAL X ARE *  ,3(2X,E12.5))
        WRITE(6 ,10)
   10    FORMAT(        18X,24HCOMPLEX PROCEDURE OF BOX)
        WRITE(6 ,18)
   18    FORMAT(   2X,10HPARAMETERS)
        WRITE(6 ,11)N,M,K,ITMAX,IC,ALPHA,BETA,GAMMA,DELTA
   11    FORMAT(   2X,4HN = ,I2,3X,4HM = ,I2,3X,4HK = ,I2,2X,8HITMAX = ,
       1 I4,2X,5HIC = ,I2,//,2X,8HALPHA = ,F5.2,5X,7HBETA = ,F10.5,3X,
       28HGAMMA = ,I2,3X,8HDELTA = ,F8.5)
        IF(IPRINT)40,50,40
  40    WRITE(6 ,12)
   12    FORMAT(//,2X,14HRANDOM NUMBERS)
        DO 200 J=2,K
        WRITE(6 ,13)(J,I,R(J,I),I=1,N)
   13    FORMAT(/,3(2X,2HR(,I2,1H,,I2,4H) = ,F8.4,2X))
  200   CONTINUE
C
```

```
   50 CALL CONSX(N,M,K,ITMAX,ALPHA,BETA,GAMMA,DELTA,X,R,F,IT,IEV2,
     1NO,G,H,XC,IPRINT)
C
      IF(IT-ITMAX) 20,20,30
   20 WRITE(6 ,14) F(IEV2)
   14  FORMAT(     2X,30HFINAL VALUE OF THE FUNCTION = ,E20,8)
      WRITE(6 ,15)
   15  FORMAT(    2X,14HFINAL X VALUES)
      DO 300 J=1,N
      WRITE(6 ,16) J,X(IEV2,J)
   16  FORMAT(/,2X,2HX(,I2,4H) = ,E20,8)
  300 CONTINUE
      CALL PUPRO(X,IEV2,IDIF)
      GO TO 999
C
   30 WRITE(6 ,17) ITMAX
   17  FORMAT(///,2X,38HTHE NUMBER OF ITERATIONS HAS EXCEEDED ,I4,10X,
     1 18HPROGRAM TERMINATED)
  999 CONTINUE
      WRITE(6,106) IT
  106 FORMAT(2X,*ITERATION NO =*,I4)
  299 CONTINUE
  399 CONTINUE
      STOP
      END
$IBFTC CONSX
      SUBROUTINE CONSX(N,M,K,ITMAX,ALPHA,BETA,GAMMA,DELTA,X,R,F,IT,
     1IEV2,NO,G,H,XC,IPRINT)
C     COORDINATES SPECIAL PURPOSE SUBROUTINES
C
C     ARGUEMENT LIST
C     IT        =ITERATION INDEX
C     IEV1      = INDEX OF POINT WITH MINIMUM FUNCTION VALUE
C     IEV2      = INDEX OF POINT WITH MAXIMUM FUNCTION VALUE
C     I         =POINT INDEX
C     KODE      = CONTROL KEY USED TO DETERMINE IF IMPLUCIT CONSTRAINTS
C     ARE PROVIDED
C     K1        = DO LOOP LIMIT
C
C     ALL OTHER PREVIOUSLY DEFINED IN MAIN LINE
C
      DIMENSION X(6,4),R(6,3),F(6),G(4),H(4),XC(3),XBEST(3)
      INTEGER GAMMA
C
      IT=1
      KODE=0
      IF(M-N)20,20,10
   10 KODE=1
   20 CONTINUE
      DO 40 II=2,K
      DO 30 J=1,N
   30 X(II,J)=0.0
   40 CONTINUE
C
C     CALCULATE COMPLEX POINTS AND CHECK AGAINST CONSTRAINTS
C
      DO 65 II=2,K
      DO 50 J=1,N
      I=II
      CALL CONST(N,M,K,X,G,H,I)
      X(II,J)=G(J)+ R(II,J)*(H(J)-G(J))
   50 CONTINUE
      K1=II
      CALL CHECK(N,M,K,X,G,H,I,KODE,XC,DELTA,K1)
      IF(II-2)51,51,55
   51 IF(IPRINT) 52,65,52
   52 WRITE(6 ,18)
```

```
   18  FORMAT(//,2X,30HCOORDINATES OF INITIAL COMPLEX)
      IO=1
      WRITE(6 ,19)(IO,J,X(IO,J),J=1,N)
   19  FORMAT(/,3(2X,2HX(,I2,1H,,I2,4H) = ,1PE13.6))
   55 IF(IPRINT)56,65,56
   56 WRITE(6 ,19) (II,J,X(II,J),J=1,N)
   65 CONTINUE
      K1=K
      DO 70 I=1,K
      CALL FUNC(N,M,K,X,F,I)
   70 CONTINUE
      KOUNT=1
      IA=0
C
C     FIND POINT WITH LOWEST FUNCTION VALUE
C
      IF(IPRINT) 72,80,72
   72 WRITE(6 ,21)
   21  FORMAT(/,2X,22HVALUES OF THE FUNCTION)
      WRITE(6 ,22)(J,F(J),J=1,K)
   22  FORMAT(/,3(2X,2HF(,I2,4H) = ,1PE13.6))
   80  IEV1=1
      DO 100 ICM=2,K
      IF(F(IEV1)-F(ICM)) 100,100,90
   90 IEV1=ICM
  100 CONTINUE
C
C     FIND POINT WITH HIGHEST FUNCTION VALUE
C
      IEV2=1
      DO 120 ICM=2,K
      IF(F(IEV2)-F(ICM)) 110,110,120
  110 IEV2=ICM
  120 CONTINUE
      DO 111 JJ=1,N
  111 XBEST(JJ)=X(IEV2,JJ)
C     CHECK CONVERGENCE CRITERIA
      IF(F(IEV2)-(F(IEV1)+BETA)) 140,130,130
  130 KOUNT=1
      GO TO 150
  140 KOUNT=KOUNT+1
      IF(KOUNT-GAMMA)150,240,240
C     REPLACE POINT WITH LOWEST FUNCTION VALUE
  150 CALL CENTR(N,M,K,IEV1,I,XC,X,K1)
      DO 160 JJ=1,N
  160 X(IEV1,JJ)=(1.0+ALPHA)*(XC(JJ))-ALPHA*(X(IEV1,JJ))
      I=IEV1
      CALL CHECK(N,M,K,X,G,H,I,KODE,XC,DELTA,K1)
      CALL FUNC(N,M,K,X,F,I)
C     REPLACE NEW POINT IF IT REPEATS AS LOWEST FUNCTION VALUE
      ICHEK=0
      IKNT=0
  170 IEV2=1
      ICHEK=ICHEK+1
      DO 190 ICM=2,K
      IF(F(IEV2)-F(ICM))190,190,180
  180 IEV2=ICM
  190 CONTINUE
      IF (IEV2-IEV1) 220,200,220
  200 DO 210 JJ=1,N
      X(IEV1,JJ)=(X(IEV1,JJ)+XC(JJ))/2.0
  210 CONTINUE
      I=IEV1
      CALL CHECK(N,M,K,X,G,H,I,KODE,XC,DELTA,K1)
      CALL FUNC(N,M,K,X,F,I)
      IF(ICHEK-25) 170,202,202
  202 IKNT=IKNT+1
```

```
      WRITE(6,203) IKNT
  203 FORMAT(2X,*IKNT= *,I3)
      IF(IKNT.GE.3) RETURN
      DO 410 JL =1,N
      X(IEV1,JL) =(X(IEV1,JL) +XBEST(JL))/2.0
  410 CONTINUE
      GO TO 170
  220 CONTINUE
      IF(IPRINT) 230,228,230
  230 PRINT 023,IT
  23  FORMAT(I2)
      PRINT 024
  024 FORMAT(/,2X,30HCOORDINATES OF CORRECTED POINT)
      PRINT 019,(IEV1,JC,X(IEV1,JC),JC=1,N)
      PRINT 021
      PRINT 022,(I,F(I),I=1,K)
      PRINT 025
  025 FORMAT(/,2X,27HCOORDINATES OF THE CENTROID)
      PRINT 026,(JC,XC(JC),JC=1,N)
  026 FORMAT(/,3(2X,2HX(,I2,6H,C) = ,E14.6,4X))
  228 IT=IT+1
      IF(IT-ITMAX)80,80,240
  240 RETURN
      END
$IBFTC CHECK
      SUBROUTINE CHECK(N,M,K,X,G,H,I,KODE,XC,DELTA,K1)
C     ARGUEMENT LIST
C     ALL ARGUEMENTS DEFINED IN MAIN LINE AND CONX
      DIMENSION X(6,4),G(4),H(4),XC(3)
   10 KT=0
      CALL CONST(N,M,K,X,G,H,I)
C     CHECK AGAINST EXPLICIT CONSTRAINTS
      DO 50 J=1,N
      IF(X(I,J)-G(J)) 20,20,30
   20 X(I,J)=G(J)+DELTA
      GO TO 50
   30 IF(H(J)-X(I,J))40,40,50
   40 X(I,J)=H(J)-DELTA
   50 CONTINUE
      IF(KODE)110,110,60
C     CHECK AGAINST IMPLICIT CONSTRIANTS
   60 NN=N+1
      DO 100 J=NN,M
      CALL CONST(N,M,K,X,G,H,I)
      IF(X(I,J)-G(J))80,70,70
   70 IF(H(J)-X(I,J)) 80,100,100
   80 IEV1=I
      KT=1
      CALL CENTR(N,M,K,IEV1,I,XC,X,K1)
      DO 90 JJ=1,N
      X(I,JJ)=(X(I,JJ)+XC(JJ))/2.0
   90 CONTINUE
  100 CONTINUE
      IF(KT)110,110,10
  110 RETURN
      END
$IBFTC CENTR
      SUBROUTINE CENTR(N,M,K,IEV1,I,XC,X,K1)
      DIMENSION X(6,4),XC(3)
      DO 20 J=1,N
      XC(J)=0.0
      DO 10 IL=1,K1
   10 XC(J)=XC(J)+X(IL,J)
      RK=K1
   20 XC(J)=(XC(J)-X(IEV1,J))/(RK-1.0)
      RETURN
      END
```

```
$IBFTC FUNC
      SUBROUTINE FUNC(N,M,K,X,F,I)
      DIMENSION X(6,4),F(6)
      COMMON THETA(4,4),P(4),OSI(4,4),DET1,DET2,DET3,DET4,
     1               A(3,3),B(2,2),C(3,3),D(2,2),E(2,2),AF(2,2), G(4,4),
     2  H(2,2)
      X1=X(I,1)
      X2=X(I,2)
      X3=X(I,3)
      X11=X1*X1
      X22=X2*X2
      X33=X3*X3
      X12=X1*X2
      X23=X2*X3
      X13=X1*X3
C     ELEMENTS OF VARIANCE COVARIANCE MATRIX OF MODEL 1FOR (N+1)RUN
      ASI11=B(1,1)+A(1,1)*X11
      ASI12=B(1,2)-A(1,1)*X11+A(1,2)*X12-A(1,3)*X13
      ASI21=ASI12
      ASI22=B(2,2)+A(1,1)*X11+A(2,2)*X22+A(3,3)*X33-2.*A(1,2)*X12
     1-2.*A(2,3)*X23+2.*A(1,3)*X13
C     ELEMENTS OF VARIANCE COVARIANCE MATRIXOF MODEL 2FOR (N+1)RUN
      BSI11=D(1,1)+C(1,1)*X11+C(2,2)*X22-2.*C(1,2)*X12
      BSI12=D(1,2)-C(1,1)*X11-(C(2,2)+C(2,3))*X22+(2.*C(1,2)+C(1,3))*X12
      BSI21=BSI12
       BSI22=D(2,2)+C(1,1)*X11+(C(2,2)+2.*C(2,3)+C(3,3))*X22-2.*(C(1,2)+
     1 C(1,3))*X12
C     ELEMENTS OF VARIANCE COVARIANCE MATRIX OF MODEL3FOR(N+1)RUN
      CSI11=E(1,1)*X11+AF(1,1)
      CSI12=AF(1,2)-E(1,1)*X11+E(1,2)*X12
      CSI21=CSI12
      CSI22=AF(2,2)+E(1,1)*X11-2.*E(1,2)*X12+E(2,2)*X22
C     ELEMENTS OF VARIANCE COVARIANCE MATRIX OF MODEL4FOR(N+1) RUN
      DSI11=H(1,1)+G(1,1)*X11-2.*G(1,2)*X12+G(2,2)*X22
      DSI12=H(1,2)-G(1,1)*X11+(2.*G(1,2)+G(1,3))*X12-(G(2,3)+G(2,2))*X22
     1-G(1,4)*X13+G(2,4)*X23
      DSI21=DSI12
      DSI22=H(2,2)+G(1,1)*X11+(G(2,2)+2.*G(2,3)+G(3,3))*X22+G(4,4)*X33-2
     1.*(G(2,4)+G(3,4))*X23+2.*G(1,4)*X13 -2.0*(G(1,2)+G(1,3))*X12
C     DETERMINANT OF THE MATRIX
      DET1=ASI11*ASI22-ASI12*ASI12
      DET2=BSI11*BSI22-BSI12*BSI12
      DET3=CSI11*CSI22-CSI12*CSI12
      DET4=DSI11*DSI22-DSI12*DSI12
C     ELEMENTS OF INVERSES OF VARIANCE COVARIANCE MATRIX OF MODEL1
      OAS11=ASI22/DET1
      OAS12=-ASI12/DET1
      OAS21=OAS12
      OAS22=ASI11/DET1
C     ELEMENTS OF INVERSES OF VARIANCE COVARIANCE MATRIX OF MODEL2
      OBS11=BSI22/DET2
      OBS12=-BSI12/DET2
      OBS21=OBS12
      OBS22=BSI11/DET2
C     ELEMENTS OF INVERSES OF VARIANCE COVARIANCE MATRIX OF MODEL3
      OCS11=CSI22/DET3
      OCS12=-CSI12/DET3
      OCS21=OCS12
      OCS22=CSI11/DET3
C     ELEMENTS OF INVERSES OF VARIANCE COVARIANCE MATRIX OF MODEL4
      ODS11=DSI22/DET4
      ODS12=-DSI12/DET4
      ODS21=ODS12
      ODS22=DSI11/DET4
      OSI(1,1)=OAS11
      OSI(1,2)=OAS12
      OSI(1,3)=OAS21
```

```
      OSI(1,4)=OAS22
      OSI(2,1)=OBS11
      OSI(2,2)=OBS12
      OSI(2,3)=OBS21
      OSI(2,4)=OBS22
      OSI(3,1)=OCS11
      OSI(3,2)=OCS12
      OSI(3,3)=OCS21
       OSI(3,4)=OCS22
       OSI(4,1)=ODS11
      OSI(4,2)=ODS12
      OSI(4,3)=ODS21
      OSI(4,4)=ODS22
C     EXPRESSION FOR Y1-Y2
      Y12=-(THETA(1,1)-THETA(2,1)) *X1-THETA(2,2)*X2
      Y21=(THETA(1,1)-THETA(2,1))*X1-(THETA(1,2)-THETA(2,2)-THETA(2,3))*
     1 X2+THETA(1,3)*X3
C     EXPRESSION FOR Y1-Y3
      Y13=-(THETA(1,1)-THETA(3,1))*X1
      Y31=(THETA(1,1)-THETA(3,1))*X1-(THETA(1,2)-THETA(3,2))*X2+THETA(1,
     13)*X3
C     EXPRESSION FOR Y1-Y4
      Y14=-(THETA(1,1)-THETA(4,1))*X1-THETA(4,2)*X2
      Y41=(THETA(1,1)-THETA(4,1))*X1-(THETA(1,2)-THETA(4,2)-THETA(4,3))
     1*X2+(THETA(1,3)-THETA(4,4))*X3
C     EXPRESSION FOR Y2-Y3
      Y23=-(THETA(2,1)-THETA(3,1))*X1+THETA(2,2)*X2
      Y32=(THETA(2,1)-THETA(3,1))*X1-(THETA(2,2)-THETA(3,2)+THETA(2,3))*
     1 X2
C     EXPRESSION FOR Y2-Y4
      Y24=-(THETA(2,1)-THETA(4,1))*X1+(THETA(2,2)-THETA(4,2))*X2
      Y42=(THETA(2,1)-THETA(4,1))*X1-(THETA(2,2)-THETA(4,2)-THETA(4,3)+T
     1 HETA(2,3))*X2-(THETA(4,4)*X3)
C     EXPRESSION FOR Y3-Y4
      Y34=-(THETA(3,1)-THETA(4,1))*X1-THETA(4,2)*X2
      Y43=(THETA(3,1)-THETA(4,1))*X1-(THETA(3,2)-THETA(4,2)-THETA(4,3))*
     1X2-THETA(4,4)*X3
C       EVALUATION OF KV
C     FOR P1 AND P2
      TRA12=ASI11*OBS11+ASI12*OBS12+BSI11*OAS11+BSI12*OAS12-2,
      TRA21=ASI12*OBS12+ASI22*OBS22+BSI12*OAS12+BSI22*OAS22-2,
      AM11=OAS11+OBS11
      AM12=OAS12+OBS12
      AM21=AM12
      AM22=OAS22+OBS22
      RES12=AM11*Y12*Y12+2,*AM12*Y12*Y21+AM22*Y21*Y21
      P12=P(1)*P(2)*(TRA12+TRA21+RES12)
C     FOR P1 AND P3
      TRA13=ASI11*OCS11+ASI12*OCS12+CSI11*OAS11+CSI12*OAS12-2,
      TRA31=ASI12*OCS12+ASI22*OCS22+CSI12*OAS12+CSI22*OAS22-2,
      BM11=OAS11+OCS11
        BM12=OAS12+OCS12
       BM22=OAS22+OCS22
      RES13=BM11*Y13*Y13+2,*BM12*Y13*Y31+BM22*Y31*Y31
      P13=P(1)*P(3)*(TRA13+TRA31+RES13)
C      FOR P1  AND P4
      TRA14=ASI11*ODS11+ASI12*ODS12+DSI11*OAS11+DSI12*OAS12-2,
      TRA41=ASI12*ODS12+ASI22*ODS22+DSI12*OAS12+DSI22*OAS22-2,
       CM11=OAS11+ODS11
       CM12=OAS12+ODS12
      CM21=CM12
      CM22=OAS22+ODS22
      RES14=CM11*Y14*Y14+2,*CM12*Y14*Y41+CM22*Y41*Y41
      P14=P(1)*P(4)*(TRA14+TRA41+RES14)
C     FOR P2 AND P3
      T[illegible]1+BS[illegible]*OCS12+CBS11*CSI11+CSI12*OBS12-2,
      [illegible]32=BSI12*OCS12+[illegible]*OCS22+CSI12*CBS12+CSI22*OBS22-2,
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      DM11=OBS11+OCS11
       DM12=OBS12+OCS12
      DM21=DM12
      DM22=OBS22+OCS22
      RES23=DM11*Y23*Y23+2.*DM12*Y23*Y32+DM22*Y32*Y32
      P23=   P(2)*P(3)*(TRA23+TRA32+RES23)
C     FOR P2 AND P4
      TRA24=BSI11*ODS11+BSI12*ODS12+DSI11*OBS11+DSI12*OBS12-2.
      TRA42=BSI12*ODS12+BSI22*ODS22+DSI12*OBS12+DSI22*OBS22-2.
      EM11=OBS11+ODS11
       EM12=OBS12+ODS12
      EM21=EM12
        EM22=OBS22+ODS22
      RES24=EM11*Y24*Y24+2.*EM12*Y24*Y42+EM22*Y42*Y42
      P24=   P(2)*P(4)*(TRA24+TRA42+RES24)
C     FOR P3 AND P4
      TRA34=CSI11*ODS11+CSI12*ODS12+DSI11*OCS11+DSI12*OCS12-2.
      TRA43=CSI12*ODS12+CSI22*ODS22+DSI12*OCS12+DSI22*OCS22-2.
      FM11=OCS11+ODS11
      FM12=OCS12+ODS12
      FM21=FM12
      FM22=OCS22+ODS22
      RES34=FM11*Y34*Y34+2.*FM12*Y34*Y43+FM22*Y43*Y43
      P34=   P(3)*P(4)*(TRA34+TRA43+RES34)
       F(I)=0.5*(P12+P13+P14+P23+P24+P34)
      RETURN
      END
$IBFTC CONST
      SUBROUTINE CONST(N,M,K,X,G,H,I)
      DIMENSION X(6,4),G(4),H(4)
      G(1)=0.
      H(1)=30.0
      G(2)=0.
      H(2)=30.0
      G(3)=0.
      H(3)=30.0
      G(4)=0.
      H(4)=90.0
      X(I,4)=X(I,1)+X(I,2)+X(I,3)
      RETURN
      END
$IBFTC POPRO
      SUBROUTINE POPRO(X,IEV2,IDIF)
      DIMENSION X(6,4),Y(4,2)
      COMMON THETA(4,4),P(4),OSI(4,4),DET1,DET2,DET3,DET4 ,
     1                 A(3,3),B(2,2),C(3,3),D(2,2),E(2,2),AF(2,2),AG(4,4),
     2 AH(2,2)
       INTEGER U
       XA=X(IEV2,1)
       XB=X(IEV2,2)
       XC=X(IEV2,3)
       U=2
       WRITE(6,20)((THETA(I,J),J=1,4),I=1,4)
       WRITE(6,20) ((OSI(I,J),J=1,4),I=1,4)
       WRITE(6,20) DET1,DET2,DET3,DET4
       WRITE(6,20) (P(I),I=1,4)
   20  FORMAT(2X,8(2X,E12.5))
C      MODEL 1  A-B=C
       Y11=-THETA(1,1)*XA
       Y13=THETA(1,2)*XB-THETA(1,3)*XC
       Y12=-Y11-Y13
       IF(IDIF.LE.5) GO TO 35
       Y(1,1)=Y11+0.6321*0.003
       Y(1,2)=Y12+0.6534*0.003
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      Y(1,2)=      (THETA(3,1)*XA-THETA(3,2)*XB) +0,6542*0,003
      Y133=      (THETA(3,2)*XB) +0,9832*0,003
  36  DIF11=Y(1,1)-Y11
      DIF12=Y(1,2)-Y12
      CON1=1,0/((DET1**0,5)*(6,28**U))
      POS1=-0,5*(OSI(1,1)*DIF11*DIF11+2,*OSI(1,2)*DIF11*DIF12+OSI(1,4)*D
     1 IF12*DIF12)
      POSP1=CON1*EXP(POS1)
C     MODEL 2  A=B-C
      Y21=-THETA(2,1)*XA+THETA(2,2)*XB
      Y23=THETA(2,3)*XB
      Y22=-Y21-Y23
      Y(2,1)=Y(1,1)
      Y(2,2)=Y(1,2)
      DIF21=Y(2,1)-Y21
      DIF22=Y(2,2)-Y22
      CON2=1,0/((DET2**0,5)*(6,28**U))
      POS2=-0,5*(OSI(2,1)*DIF21*DIF21+2,*OSI(2,2)*DIF21*DIF22+OSI(2,4)*D
     1 IF22*DIF22)
      POSP2=CON2*EXP(POS2)
C     MODEL 3  A-B-C
      Y31=-THETA(3,1)*XA
      Y33=THETA(3,2)*XB
      Y32=-Y31-Y33
      Y(3,1)=Y(1,1)
      Y(3,2)=Y(1,2)
      DIF31=Y(3,1)-Y31
      DIF32=Y(3,2)-Y32
      CON3=1,0/((DET3**0,5)*(6,28**U))
      POS3=-0,5*(OSI(3,1)*DIF31*DIF31+2,*OSI(3,2)*DIF31*DIF32+OSI(3,4)*D
     1 IF32*DIF32)
      POSP3=CON3*EXP(POS3)
C      MODEL 4  A=B=C
      Y41=-THETA(4,1)*XA+THETA(4,2)*XB
      Y43=THETA(4,3)*XB-THETA(4,4)*XC
      Y42=-Y41-Y43
      Y(4,1)=Y(1,1)
      Y(4,2)=Y(1,2)
      DIF41=Y(4,1)-Y41
      DIF42=Y(4,2)-Y42
      CON4=1,0/((DET4**0,5)*(6,28**U))
      POS4=-0,5*(OSI(4,1)*DIF41*DIF41+2,*OSI(4,2)*DIF41*DIF42+OSI(4,4)*D
     1IF42*DIF42)
      POSP4=CON4*EXP(POS4)
      DENOM=P(1)*POSP1+P(2)*POSP2+P(3)*POSP3+P(4)*POSP4
      PROB1=P(1)*POSP1/DENOM
      PROB2=P(2)*POSP2/DENOM
      PROB3=P(3)*POSP3/DENOM
      PROB4=P(4)*POSP4/DENOM
      WRITE(6,21) XA,XB,XC,Y(1,2),Y133
  22  FORMAT(5F7,3)
  21  FORMAT(5(5X,E12,5))
      WRITE(6,210) PROB1,PROB2,PROB3,PROB4
 210  FORMAT(15X,*PROB1=*,F8,5,*PROB2=*,F8,5,*PROB3=*,F8,5,*PROB4=*,F8,5
     1 )
      RETURN
      END
$ENTRY
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